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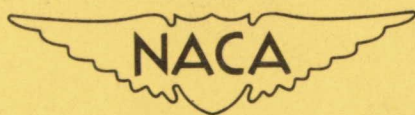
**NATIONAL ADVISORY COMMITTEE  
FOR AERONAUTICS**

**TECHNICAL NOTE 2293**

THE PHYSICAL PROPERTIES OF ACTIVE NITROGEN  
IN LOW-DENSITY FLOW

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SUMMARY

The physical properties of active nitrogen have been investigated experimentally in low-density flow at velocities extending into the supersonic region.

Measurements were made of the viscosity, the heat released during decay of the afterglow, and the free electron density in the stream. The results indicate that active nitrogen may be used for observations of flow at low density without introducing excessive disturbances in the stream, although the effects of ionization may be marked unless suitable ion traps are provided in the stream.

A compression shock wave caused an abrupt rise in the intensity of the afterglow that was followed by a further rise especially noticeable in the yellow portion of the spectrum. The intensity in the green band increased abruptly at the shock wave and remained almost constant at a value that was proportional to the increase in density of the nitrogen caused by the shock.

Experimental methods are described which appear to be useful for the study of supersonic flow at low density by the use of the Lewis-Rayleigh afterglow in shock tubes. The methods are likewise applicable to a more detailed analysis of the Lewis-Rayleigh afterglow and its transient response throughout the spectrum to the step-function changes associated with a shock wave.

The experimental results are discussed briefly to show that they are inconsistent with Mitra's theory of the afterglow and that a metastable molecular state having energies adjoining those of the B state appears to be required to explain the response of the afterglow to a shock wave.

## INTRODUCTION

The present investigation developed from a preliminary investigation (reference 1) of the characteristics of nitrogen afterglow in a supersonic stream of active nitrogen. It was found that the afterglow increased sharply in brightness when the active nitrogen was suddenly compressed in passing through a stationary shock wave. A comparison of the afterglow patterns with schlieren photographs of the flow showed that the rise in brightness of the glow coincided with the position of the shock wave. This response of the afterglow to shock waves appeared to have useful applications in visualizing flow patterns at low densities where schlieren, shadowgraph, and interferometer methods are of little or no value.

Before the active nitrogen could be used in aerodynamic applications with confidence, it appeared necessary to determine the magnitude of the effects of a shock wave and to examine the aerodynamic disturbances introduced into a stream tube by the activity that is associated with the afterglow. The various theories of the afterglow disagree widely in predicting the effect of sudden pressure changes. They are also in disagreement as to the disturbances to be expected in a stream as the result of ionization and the release of heat during decay of the afterglow.

Active nitrogen is a form of nitrogen that results from suitable excitation by a strong electric discharge at low pressures. The visible afterglow that persists after the discharge is stopped was studied by P. Lewis as early as 1900 (reference 2). The second Lord Rayleigh (R. J. Strutt) observed that the excited gas is chemically active (reference 3). He also observed that when any one of certain other elements, for example, sodium or mercury, is introduced into a stream of active nitrogen, the yellow afterglow is replaced by emission characteristic of the element added to the stream. A striking feature of the spectra of elements added to active nitrogen is that they are limited to selected members of complete spectra and are representative of low-temperature emission.

The spectrum of the nitrogen afterglow was found by Fowler and Strutt (reference 4) to include, as a characteristic feature, three bands, red, yellow, and green, that are identical with some of the bands in the first positive system of the normal nitrogen molecule. Among the many band systems of molecular nitrogen, the following may be mentioned in relation to the afterglow of active nitrogen (see energy-level diagram in fig. 1):

1. First positive system arising from transitions from B to A states
2. Second positive system, C to B
3. Fourth positive system, D to B
4. Vegard-Kaplan system involving forbidden transitions from the metastable A state to the ground state X

The spectrum of the afterglow of active nitrogen is markedly different from that of the glow discharge. The glow discharge of nitrogen includes many band systems while the spectrum of the pure afterglow of active nitrogen is restricted to the first positive system. Furthermore, certain members of the first positive system are selectively enhanced in the afterglow to produce an intensity distribution different from that of the first positive system in the nitrogen glow discharge.

Mitra has summarized in reference 5 the large amount of experimental data that have been obtained on the chemical and physical properties of active nitrogen up to 1945. Reference 5 also includes an extensive bibliography of 102 entries relating to active nitrogen. It is clearly established that the yellow afterglow characteristic of active nitrogen contains no atomic lines and contains no band system which cannot be ascribed to the normal nitrogen molecule. Kaplan, reference 6, has named the yellow afterglow of active nitrogen the "Lewis-Rayleigh" afterglow to distinguish it from other afterglows in nitrogen. The emission of the afterglow takes place by allowed transitions between electronic states of the molecule; therefore, the long lifetime of the afterglow is not to be attributed to the persistence of excited states of the upper levels from which radiating transitions occur.

Extensive experiments were carried out by Rayleigh (references 3, 7, and 8) to determine many of the chemical and physical properties of active nitrogen. He gave special attention to the visual brightness of the afterglow in vessels arranged for changing the volume, pressure, and temperature of the excited gas. He also measured the energy transferred to metals placed in the excited gas and investigated the ionization associated with active nitrogen.

Different theories have been proposed during the past 40 years to provide a model that would account for the energy of the active centers and the long life of the afterglow.

Some of the theories assume that the active centers are nitrogen atoms that recombine slowly under conditions that produce the afterglow. The formulation of atomic models of active nitrogen has been open to

question because the dissociation energy of nitrogen has not been definitely established within the probable range of 7.3 to 9.5 electron volts.

Theories that assume the active centers to be metastable molecules readily account for the energy as determined spectroscopically. However, Mitra (reference 5) has pointed out that metastable molecules do not account for the energy of 12.9 electron volts that were measured by Rayleigh in experiments on the heating of metal foil placed in active nitrogen. Mitra has proposed another theory in which he assumes the active centers to be singly ionized nitrogen molecules having the ionization energy of 15.58 electron volts. Other theories have been proposed in which it is assumed that both atomic nitrogen and metastable molecules are present in active nitrogen. The following list of postulates illustrates the wide differences among some of the theories that have been proposed:

1. The postulate of a three-body collision proposed by Cario and Kaplan (reference 9) in which two metastable nitrogen atoms of specified states and a metastable nitrogen molecule in the A electronic state collide to form from the two atoms a molecule in the B state and also raise the energy of the colliding A state molecule from the A to the B state. Both molecules may then emit the afterglow by immediate transition from the B to the A state.

2. The postulate that metastable molecules in some unspecified excited levels having an energy of approximately 9.7 electron volts undergo radiationless transition to adjacent vibrational levels of the B state from which the afterglow is emitted by allowed transition to the metastable A state (reference 10 and pp. 124 and 159 of reference 11). Other theories postulating metastable molecules are reviewed by Mitra in reference 5.

3. The postulate by Mitra that  $N_2^+$  ions persist for a long time in the active nitrogen and are neutralized in a three-body collision resulting in molecules excited to the B state from which emission takes place by allowed transition to the A state (reference 5).

The experimental work reported herein was carried out at the Langley Aeronautical Laboratory by making use of large rates of flow of active nitrogen. Measurements were made of viscosity, average energy content, ionization in the stream, and the transient response of the afterglow in different spectral regions to rapid expansion in a nozzle and to sudden compression through shock waves. Static pressures for steady flow of the active nitrogen ranged from about 0.2 to 20 millimeters of mercury. The effects of rapid pressure changes on brightness in different parts of the spectrum were observed for compressions up to 10 times the static pressure.

The evaluation of the physical properties of streaming active nitrogen provides new information for examining the various theories of the afterglow. The results of the tests also clarify some of the more important questions that arise in applications of the afterglow to studies in compressible flow at low densities.

This report, with modifications as to some details, was submitted to the University of Tennessee in the form of a thesis in partial fulfillment of the requirements for the degree of doctor of philosophy. Grateful acknowledgement is made to Dr. John D. Trimmer of the University of Tennessee, Department of Physics, for helpful discussions and suggestions during the investigation. Acknowledgement is also made to Dr. Joseph Kaplan of the University of California, Los Angeles, Calif., who first suggested that the Lewis-Rayleigh afterglow might be useful in the visualization of low-density flow.

#### POPULATION AND AVERAGE ENERGY OF ACTIVE CENTERS

Estimates from Rayleigh's measurements.- Early measurements by Rayleigh (reference 3) of the absorption of active nitrogen by phosphorous indicated that about one part in 210 of streaming nitrogen became activated by an electric discharge. Subsequent measurements by him (reference 12) of the formation of  $N_2O_3$  from nitric oxide and active nitrogen indicated one part in 40 was activated.

In a later experiment, Rayleigh (reference 8, I) measured the visible flux from a known mass of nitrogen when excited to produce as much afterglow as could readily be obtained. He found that the total flux in the visible region was roughly equivalent to the emission of one quantum for each 1,300 molecules present. Additional emission is known to occur in the infrared and ultraviolet regions so that it is to be expected that more than 1 molecule in 1,300 is activated.

Rayleigh's measurements of the heating of gold foils (reference 8, II) indicated that an extraordinary amount of energy could be recovered from the active nitrogen. He recovered in the form of heat on a piece of gold sheet as much as 12.9 electron volts of energy for each molecule that passed through the exciter. As Rayleigh pointed out, this amount of energy exceeds that available from all the gas that passes through the apparatus due to dissociation in the exciter and recombination on the gold sheet. If the gas were almost completely ionized as postulated by Mitra, there would be enough energy available from recombination on the metal surface. More recent experiments by Rayleigh (reference 12) resulted in still larger amounts of energy recovered on metal wires.

Calorimeter measurements of heat released by active nitrogen.- A stream of nitrogen was excited as described in appendix A. The active nitrogen was passed through a calorimeter as described in appendix B to measure the heat released from the stream. The intensity of the glow and the pressure were measured at the intake and the exhaust ends of the

calorimeter. The afterglow was found to decay to approximately 1/20 in passing through the calorimeter during a representative test run; this decay indicated that the excitation responsible for the afterglow also decayed to a small fraction of its initial value. It is to be expected then that the heat recovered by the calorimeter during the decay of the afterglow would be indicative of the energy carried in latent form by the active nitrogen. This expectation is supported by spectroscopic studies reported by Kaplan (reference 13) indicating that the spectrum of the Lewis-Rayleigh afterglow does not appear to change in distribution of intensity as it decays.

The results obtained in two different runs with the calorimeter are as follows (the first run was with a higher pressure in the exciter tube and the afterglow was not as bright as in the second run where the pressure was adjusted to obtain maximum intensity; presumably, the nitrogen was more highly activated in the second run):

	<u>Run 1</u>	<u>Run 2</u>
Pressure at intake, millimeters of mercury . . . . .	22.8	10.8
Pressure at exhaust, millimeters of mercury . . . . .	20.4	3.7
Mass flow, grams per minute . . . . .	2.27	2.27
Duration of run, minutes . . . . .	50	41
Temperature drop in stream as indicated by thermocouples at intake and exhaust, °C . . . . .	5	14
Temperature rise of calorimeter, °C . . . . .	0.216	0.73
Mass of water in calorimeter plus water equivalent of coil and cup, grams . . . . .	2203	2203
Heat flow to calorimeter, cal/min . . . . .	9.5	38.5
Heat flow indicated by temperature drop of stream, cal/min . . . . .	2.8	8.0
Net flow of heat ascribed to decay of active nitrogen in the calorimeter, cal/min . . . . .	6.7	30.5

Both runs were made with a grounded aluminum elbow upstream of the calorimeter to limit the flow of electrically charged particles into the calorimeter.

A subsequent run was made with the conditions the same as in run 2 except that the coil of the calorimeter was grounded and no other portion of the system was grounded between calorimeter and exciter tube. A net flow of 36.6 calories per minute was obtained in this last run, showing that the heat flow was less with the elbow grounded. The difference represents, at least in part, heating effects of the ion currents to ground.

Rayleigh's observation that copper was especially effective in quenching the afterglow suggested that a coil of copper tubing in the



calorimeter might recover more energy from the active nitrogen than one of aluminum. The glow was faintly visible in the exhaust from an aluminum coil, but no glow could be detected in the exhaust from the copper. The results of the run with a copper coil when both the aluminum elbow and the copper coil were grounded were as follows:

Pressure at intake, millimeters of mercury . . . . .	9.7
Pressure at exhaust, millimeters of mercury . . . . .	2.9
Mass flow, grams per minute . . . . .	2.27
Duration of run, minutes . . . . .	54
Temperature drop in stream, °C . . . . .	3
Temperature rise in calorimeter, °C . . . . .	1.31
Mass of water plus water equivalent of calorimeter, grams . . . . .	2159
Heat flow to calorimeter, cal/min . . . . .	52.3
Heat flow indicated by temperature drop of stream . . . . .	1.7
Net flow ascribed to decay of active nitrogen, cal/min . . . . .	50.6

As a check on the effect of the fluid friction and expansion of the gas in the coil, a run was made with the flow adjusted as usual, but with no electrical excitation of the nitrogen. In 1 hour, the temperature of the calorimeter changed less than  $0.05^{\circ}\text{C}$ , indicating an effect of only about 5 percent of the rise due to the active nitrogen.

The foregoing results show that the heat recovery is measurably increased by ion currents to the coil and by more effective quenching of the glow when copper is used instead of aluminum.

The largest amount of energy recovery, 50.6 calories per minute with the grounded copper coil, may be used in an estimate of the maximum average energy release to be expected under the conditions of the experiment. This is equivalent to 22.3 calories per gram which, if released adiabatically in a stream tube, would raise the static temperature of the stream by  $90^{\circ}\text{C}$ . This value is also equivalent to 0.027 electron volts per molecule of nitrogen. This is only about  $1/400$  the maximum value measured by Rayleigh in his first series of measurements of heating effects.

It is recognized that the present measurements were made at a pressure about 20 times that used by Rayleigh and that he observed larger heating effects per molecule of gas as the pressure was lowered. He also observed that the heating effect was decreased as he increased the distance from the gold sheet to the exciter over the range from 19 to 27 centimeters. He used a mass rate of flow only 0.075 grams per minute through a tube 2 centimeters in diameter with a static pressure of about 0.5 millimeter. This rate may be compared qualitatively with the present experiments using 2.27 grams per minute at 10-millimeter pressure through a tube having a diameter of about 2 centimeters at the exciter reducing to 1 centimeter at the calorimeter. The actual

velocities depend upon the temperature in the two experiments. As a rough approximation, the temperatures may be assumed equal, resulting in the velocity in the present experiment being about 20 times that of Rayleigh's experiment. The entrance to the calorimeter coil was about 60 centimeters from the exciter tube. The transit time of the gas from the exciter to the measuring region was probably about  $1/4$  to  $1/2$  second in Rayleigh's measurement and about  $1/10$  second in the present tests. Thus, it appears that differences in the delay in transit do not account for the calorimeter measurements' showing a lower energy of the active centers than Rayleigh measured. The present calorimeter measurements, then, disagree with the values of the energy obtained by Rayleigh in the heating of metal sheets exposed to active nitrogen.

The excitation of active nitrogen in the present tests may be compared with the excitation employed by Rayleigh, on the assumption that the intensity of the afterglow at any instant is proportional to the latent energy remaining in the excited gas. Rayleigh measured the maximum intensity of the visible portion of the afterglow under conditions that produced the brightest afterglow per unit mass of nitrogen. He found a maximum emission of approximately 504 candle seconds of flux per gram of nitrogen integrated over the lifetime of the glow. He estimated the maximum intensity immediately after the exciting discharge to be 168 candle power per gram. This is equivalent to the emission of 1 quantum per second at a wavelength of 5560 angstrom units for 1 molecule out of each 3900 molecules present.

In the present tests, measurements with an RCA 929 photocell showed the intensity at the entrance to the calorimeter to be about 85 candle power per gram of nitrogen in the stream. The 929 photocell has a spectral response designated by the manufacturer as a type "S-4," with a maximum sensitivity at about 5000 angstrom units. The photocell was calibrated by use of a standard incandescent lamp bulb. Differences between the present method and Rayleigh's method of matching the afterglow intensity with a fluorescent source employing uranium, do not allow accurate quantitative comparisons. Neither method is useful in the ultraviolet or infrared parts of the spectrum and, consequently, they measure only a portion of the total radiation.

In an effort to examine the heating effects observed by Rayleigh, a piece of gold foil 1.2 centimeters square by 0.1 millimeter was placed in the stream of active nitrogen. It was located about 20 centimeters from the nearest of the exciting discharges. No visible incandescence due to active nitrogen was obtained at any pressure or flow rate available with the apparatus on hand. Then the foil was carefully cleaned and raised to incandescence by using it as one electrode of a high-voltage discharge. As soon as the discharge to the gold was stopped, the incandescence faded out even though the exciter discharge remained in operation and the afterglow continued bright. No further effort was

made to duplicate the heating effect observed by Rayleigh since it appeared that the effect was not an essential characteristic of a stream having a bright afterglow.

The calorimeter measurements in these tests show that the population of active centers is very much less than the indications obtained by Rayleigh in his experiments on the heating of gold sheets. The heating effects observed by Rayleigh may have been largely due to heating of the gold sheet by cathode rays as pointed out by Gaydon (p. 160 of reference 11). The calorimeter measurements, then, are considered to be a more reliable indication of the population of active centers.

From the observed heat recovery of 22.3 calories per gram with a grounded copper coil, an average value of 0.023 electron volt is indicated per molecule in the stream. If the value of 9.6 electron volts is taken as the required excitation to produce afterglow, the indication is that the fraction of molecules excited to this level is of the order of one part in 240. This ratio may be compared with Rayleigh's estimate of one part in 40 from chemical activity and one part in 1300 (p. 5 of reference 8, I) from the intensity of the visible afterglow. Differences in the amount of excitation are to be expected in the different experiments. In the present investigation, the population of active centers that was obtained is not considered to be the maximum possible. Rayleigh considered that the afterglow obtained by him with much lower rates of flow approached the maximum that could reasonably be expected.

#### VISCOSITY OF ACTIVE NITROGEN

Mitra's theory in which he postulates the presence of long-lived positive molecular ions was presented in part to account for the value of 12.9 electron volts obtained by Rayleigh in heating effects on gold sheet. If this view is correct, a stream of active nitrogen would have an extraordinarily high concentration of ions. In the extreme case cited by Rayleigh, more than 80 percent of the molecules would be ionized. From data on the mobility of ions, it is to be expected that the presence of a large fraction of molecular ions would decrease strongly the viscosity of the gas.

The possibility that the viscosity of active nitrogen differed from that of normal nitrogen was investigated experimentally by measurement of the pressure drop across the calorimeter as described in appendix B. Nitrogen was pumped through the exciter and the calorimeter at a constant rate to observe whether any change in viscosity occurred when the exciter was turned on or off. Photoelectric measurements of the brightness of the afterglow indicated that the intensity per unit mass of active nitrogen was at least one-half as great as obtained

by Rayleigh. The measurements showed no change in viscosity with excitation on and off. The apparatus was sufficiently sensitive to detect a change as small as one-half of 1 percent of the absolute viscosity. These results provide no quantitative measurement of the population of active centers. The indication is strong that positive molecular ions were not present in the concentrations inferred by Mitra from Rayleigh's measurements of an average energy of 12.9 electron volts per molecule.

### ELECTRICAL PROPERTIES OF ACTIVE NITROGEN

Ionization.- Probe measurements of ionization by Rayleigh and others are conveniently summarized in reference 5, page 46 wherein Mitra concludes that:

"1. Ionization is always associated with the glow; the decrease of one is accompanied by decrease of the other. The rates are, however, not the same. The glow intensity decreases more rapidly than the ionization.

"2. The glow and the ionization persist even after the glowing gas from the discharge tube has been drawn through the space between a pair of electrodes across which a saturation current is flowing. (This is done with a view to "trap" the ions which may otherwise be expected to find their way from the discharge tube to the afterglow tube.)

"3. The ionization current between a pair of electrodes is nearly independent of the nature of metal surface. There is exception in the case of copper oxide for which the ionization current is perceptibly higher. (Copper oxide is also highly destructive of the glow.)

"4. The saturation current flowing between a pair of electrodes immersed in the glowing gas is proportional to the area of the negative electrode.

"5. The ionization is not due to emission of electrons from the surface of the metallic electrodes or to photoionization in the volume of the gas.

"6. Introduction of neutral  $N_2$  into the glowing gas increases the ionization current."

Mitra discusses ionization measurements in relation to his postulate that the walls of a vessel containing active nitrogen become conditioned in some way - "so that only a very small fraction of the impinging electrons attach themselves to the surface to form surface change."

From this viewpoint is developed (p. 65, reference 5) the conclusion that probes in the active nitrogen will absorb only a small fraction of electrons and ions that strike them and hence that probe measurements lead to too small an estimate of the ionization present in active nitrogen.

The results of an experiment by Jackson and Broadway (reference 14) appear to be in disagreement with Mitra's theory that the active centers are charged particles. They applied the Stern-Gerlach method to a study of active nitrogen. A narrow beam of active nitrogen was passed through a nonhomogeneous magnetic field to measure the magnetic moment of the active particles. The active nitrogen was detected by its effect on silver nitrate after its passage as a small jet through the field. A visible trace was produced in 2 to 5 hours with the field off. With the field on for a run of equal length, a normal Stern-Gerlach pattern was observed with no evidence being noted that the active particles carried an electrical charge. From the description of their apparatus and method, it appears that if the active nitrogen consisted of charged particles as proposed by Mitra, the pattern would have been conspicuously deflected and distorted.

Corona discharges in the stream.- In the preliminary investigation of afterglow reported in reference 1, corona discharges were observed in certain regions of the flow. The corona discharges have been observed in regions near metallic surfaces and where rapid expansion of the flow causes a large drop in pressure. Figure 2, is a photograph of the flow of active nitrogen over a double wedge at a Mach number of 2.6 which was obtained during the investigation described in reference 1. The corona discharge is to be seen in the region of the trailing edge. The corona disappeared if the flow upstream of the nozzle passed through a grounded length of electrically conducting pipe or through a grounded wire mesh.

The space discharge that occurs in the nozzle without a metallic model is shown in figure 3. Figure 3(a) is a photograph of the afterglow with no corona in the stream. The flow is directed from left to right. The patterns downstream of the minimum section reveal the dimmer regions of low pressure separated by brighter regions of high pressure resulting from waves being reflected from the boundaries of the stream. Figure 3(b) is a photograph of the flow under the same conditions except that the ground was disconnected and the space discharge occurs at several places in the nozzle. Figure 3(a) was exposed to show the afterglow patterns in the expanding section of the nozzle. It will be observed that the brighter area in the upstream end of the nozzle is over-exposed. Figure 3(b) was exposed a shorter time to show the corona patterns which are much brighter than the afterglow. This difference in exposure is evident from a comparison of the upstream end of the nozzle in the two photographs. Photoelectric measurements of the intensity of the afterglow in the regions between the screen and the space discharge

showed no change when the screen was grounded to remove the discharge. With the space discharge localized in the nozzle, the intensity of the afterglow in the exhaust line was less with the discharge taking place than when the discharge was prevented by grounding the screen. This is in agreement with the observation by Kaplan that a weak electrodeless discharge in active nitrogen, such as that caused by a Tesla coil, quenches the afterglow.

Consideration has been given to the possibility that the space discharge illustrated in figure 3(b) may have resulted from an electrical discharge between the exciter system and the ground. When a magnetic field was applied across the stream at the corona, the discharge was conspicuously deflected in a manner to be expected of negative ions moving with the stream. Since the exciter was powered by alternating current and the deflection was unidirectional, the space discharge in the stream appears not to be the result of a discharge current from exciter to ground.

The observation that removal of ions from the stream had no effect on the afterglow was further confirmed by comparing the spectrum of the glow with and without the screen grounded. The spectrum of the afterglow obtained with a small quartz spectrograph (Gaertner model L234-150) on Eastman 103-G plates showed no difference in distribution of intensity or exposure with the screen grounded and ungrounded. Thus, it appears that the presence of ions in the stream may vary widely without affecting the afterglow except that, if the space charge becomes sufficient to cause a discharge, the afterglow is quenched.

Microwave measurements.- In view of the questions that have been raised, especially by Mitra's theory, regarding the data on ion density obtained with probes, measurements of electron density were carried out by the use of a resonant cavity as described in appendix C. The electron density measured in this way depended upon the distance from exciter to the resonant cavity and whether or not there was a grounded length of pipe ahead of the cavity. With the cavity only about 12 inches from the nearest of the three exciting discharges, the effects were too erratic to allow a reading of the resonant wavelength. When an intervening length of 12 inches of aluminum tubing between exciter and cavity was grounded, the resonant peak of the cavity was easily read, showing a resonant frequency of 2,601.87 megacycles with the exciter on and the nitrogen at a pressure of 8.0 millimeters. With the exciter off, the resonant frequency was 2,601.15 megacycles. The shift of 0.73 megacycles represented an electron density of  $7.7 \times 10^8$  free electrons per cubic centimeter. It is estimated from thermocouple measurements of temperature of the stream in a similar arrangement that the temperature was roughly 400° K from which it is estimated that the stream density was  $1.9 \times 10^{17}$  molecules per cubic centimeter. The shift in resonant

frequency thus indicates an electron density of about one free electron for each  $2.5 \times 10^8$  molecules in the stream. Additional readings of the shift in resonant frequency are listed in table I for several pressures.

The results for runs 1 to 7 in table I show frequency shifts that were too small for accurate measurements. Runs 8 to 14 show that the free electron density increased as the pressure increased up to 12 millimeters of mercury and decreased at higher pressures. The free electron density was observed to decrease but did not become zero when a conducting length of pipe was grounded.

It is of interest to compare the measured values of the free electron density in active nitrogen with values measured by similar methods in another gas. The free electron density in active nitrogen was found to be as high as  $1.89 \times 10^9$  per cubic centimeter at a pressure of 12 millimeters of mercury. Reference 15 reports a value of the order of  $10^{10}$  per cubic centimeter in neon at a time of about 750 microseconds after the end of an electric discharge. At the instant of their measurement, the neon was glowing due to radiative recombination in the plasma. This comparison shows that the free electron density in active nitrogen is notably high and it is not surprising that space discharges are observed in a stream having this large a concentration of free electrons. Even so, the concentration is only about 1 free electron for each  $2.3 \times 10^8$  molecules of nitrogen in the stream, which is much less than the population of active centers indicated by the calorimeter measurements.

Effects of a magnetic field on measurements in the stream.- Brief observations were made of the ion currents obtainable with aluminum electrodes inserted in the stream of active nitrogen. The electrodes were flat cylindrical disks, 1/4 inch in diameter and 1/8 inch thick with embedded thermocouples of iron constantan. The ion currents were measured by means of a direct-current microammeter in the arrangement illustrated in figure 4. With the electrodes approximately flush with the walls of the glass tubing through which the active nitrogen flowed, no current was measured when the circuit was insulated from the ground. It was found that a current of several microamperes could be obtained when a magnetic field of a few hundred gauss was applied transverse to both the stream and a line joining the electrodes. A corona discharge was also observed in the vicinity of one electrode but not the other. The corona shifted from one to the other and the direction of the current reversed when the magnetic field was reversed. The corona appeared always at the electrode toward which streaming electrons would be deflected by the magnetic field. The thermocouples showed this electrode to be of the order of  $10^{\circ}$  C warmer than the opposite one.

Effects similar to that caused by the magnetic field were observed when one electrode was moved to the center of the stream and the other withdrawn about 1/4 inch into the side arm. The current obtained in this way was directed as if negative ions were being collected from the stream. The ion current obtainable with the magnetic field diminished but did not become zero when a section of the pipe through which the active nitrogen flowed was grounded upstream of the electrodes. Microwave measurements at the same time with the resonant cavity showed that electron density in the stream a few inches ahead of the pair of electrodes also decreased when the pipe was grounded. The decrease indicated by the microwave measurement was roughly proportional to the decrease in the ion current measured between the electrodes.

These results are to be expected, and they indicate that the probes may be used in active nitrogen in a qualitative way. In contrast, Mitra's theory assumes that active nitrogen conditions the electrode surfaces in such a way as to reflect electrons and allow free access of the positive ions to the electrodes. From this, Mitra inferred that electrodes in active nitrogen measured more current due to positive ions than to free electrons. His view disagrees with the present results which indicate that the electrodes respond to negative rather than positive charges in the stream.

## TRANSIENT RESPONSE OF AFTERGLOW TO PRESSURE CHANGES

### IN A NOZZLE

The preliminary tests (reference 1) of active nitrogen in supersonic flow were successful in demonstrating that the afterglow responded without apparent delay to the sudden compression associated with a shock wave. Figures 5(a) and 5(b) are photographs from reference 1 of the flow about a double wedge showing that the position of a shock wave in the schlieren photograph (fig. 5(a)) coincides within experimental accuracy with the discontinuous rise in brightness of the afterglow at the shock wave in figure 5(b).

The various theories of the afterglow differ as to the transient response of the intensity to be expected from the almost discontinuous changes in pressure, density, temperature, and collision frequency that are associated with supersonic flow and shock waves. A number of experiments by Rayleigh and others on the effects of pressure are reviewed by Mitra (reference 5) to determine whether the process leading to the afterglow is merely a monomolecular decay or requires two-body or three-body collisions. He concluded that the rate of the process varied as the cube of the pressure, indicating that a three-body collision transferred energy from the long-lived state to the state from which the afterglow was emitted.



The experiments carried out by Rayleigh involved measurements of the total intensity by visual or photoelectric comparison with a standard source. His method allowed measurements of the steady state established after the pressure change in vessels. It did not allow an examination of the transient characteristics within the short time interval that may be required to reestablish equilibrium after a change. The present methods represent a new approach in that use is made of supersonic flow to allow an examination of the time history of the effects of abrupt changes in pressure and density.

The response of the afterglow to flow through a stationary shock in a nozzle was observed in reference 1 to begin sharply and discontinuously with no delay that could be seen by comparison of the schlieren and afterglow patterns. The time delay that could have been detected may be estimated roughly from the velocity of the gas through the nozzle. At the Mach number of 2.6 and stagnation temperature of 325° K, the stream velocity ahead of a shock wave is 480 millimeters per millisecond. It is estimated that a displacement of less than 5 millimeters could readily be observed in comparing the position of the shock wave in the schlieren photograph with that in the afterglow pattern. In this way, a delay of as much as 1/100 millisecond in the initial response of the afterglow to a pressure change would have been detected in the preliminary investigation of reference 1.

An additional feature of the present methods is that the transient response of the afterglow may be examined in different regions of the spectrum. In studying the effects of an expansion, an image of the minimum section of a supersonic nozzle was focused on the center of the entrance slit of a small quartz spectrograph with the stream directed parallel to the slit. One end of the slit formed a spectrum of the afterglow in the settling chamber. The spectrum at the other end of the slit was from the much lower pressure region in the expanding section of the nozzle.

Figure 6 is a spectrogram of the afterglow in the nozzle. The flow was parallel to the slit of the spectrograph and directed from top to bottom of the figure. The slit was opened rather wide because of the low intensity of the afterglow. The upstream end of the slit was exposed 2 hours and the downstream end, where the flow expands to a low pressure, was exposed 4 hours. The dashed line in the figure and the break in the spectrum lines indicate the position of the minimum section of the nozzle. The plate was an Eastman I-N which is sensitive to the near infrared but not very sensitive to the green and blue.

The different bands of the afterglow are shown distinctly except for the green, but the vibrational structure is not resolved. It is seen that the relative intensities of the different bands do not change

as the gas expands from 8 millimeters of mercury pressure at one end to less than 2 at the other end of the slit. It should be pointed out that the time interval required for the gas to pass across the slit of the spectrograph is of the order of  $1/10$  millisecond and it is possible that some redistribution of spectral intensity in response to the drop in pressure could occur over a longer time. The inverse change, a compression, did show a redistribution as described in the paragraphs to follow.

#### MEASUREMENTS IN A SHOCK TUBE

Pressure and intensity.- Efforts were made to determine the variation of intensity of the afterglow with pressure from measurements in a nozzle. Satisfactory results were not obtained because of the cross-stream gradients in the flow and the apparent quenching near the walls. Instead of undertaking to develop a satisfactory nozzle, it appeared more promising to make use of shock tubes in the study of transient phenomena associated with shock waves.

Appendix D describes the shock tube and the associated instruments that were used in measuring the intensity and pressure changes caused by a normal shock. Reference 16 is a review of the characteristics of shock tubes as used in studying transient and quasi-steady flow.

The records of the photocell used in measuring the intensity showed, in agreement with observations in the nozzle, that a shock wave was accompanied by an abrupt rise in brightness of the afterglow on the high-pressure side of the shock. A record of the time history of the photoelectric response and the signal from the pressure gage are included as figure 7 for five different pressures. The response rate of the photocell to the afterglow may be compared with its response to a step function light pulse shown in figure 8 which is a record taken with a light chopper producing practically square wave pulses.

The time constant of the photocell and associated apparatus is shown by an examination of figure 8 to be less than  $1/10$  millisecond. This may be compared with the time required for a shock wave to pass the window of the photocell. The window was not more than 3 millimeters wide for any of the tests. Wave velocities were measured relative to the photocell and found to be from 500 to 1000 millimeters per millisecond, depending upon the strength of the shock and the stream velocity. From this, the time for the shock to travel across the photocell is found to be less than  $1/100$  millisecond and the record from the photocell would be expected to show a step function change when a shock wave passes.

The gage and its circuits, which are described in appendix D, have a flat response up to about 5000 cycles per second. The response is down 10 decibels at 10,000 cycles per second. The pressure recorder has a lag in response of about 1/10 millisecond that causes a time displacement attributable to the filter circuits. This lag accounts for the difference in times of initial rise in the records of pressure and light intensity in figure 7.

The record in figure 7(a) shows the pressure jump across the shock wave to be practically a step function with a plateau that is flat for about 1 millisecond after the shock wave passed the gage. The rise in the light intensity is very steep at first and then more gradual up to a peak value 0.7 millisecond after the shock wave passed. After that time, the intensity dies away as the mixing zone between active nitrogen and the inflowing air passes the window.

The rise in light intensity that results from the compression at a shock wave may be evaluated from the records by determining the ratio of the intensity ahead of the shock to that behind the shock. This ratio may be determined by measuring the rise in the output of the photocell as the shock passes, and the subsequent drop when the afterglow is extinguished after the mixing zone passes. The initial intensity,  $I_1$ , is the difference between the output before the shock wave arrives and the zero level to which it drops after the mixing zone passes. The transient increase,  $\Delta I$ , caused by the shock is the rise above the initial intensity  $I_1$ . The transient intensity,  $I_2$ , after the shock is the sum of  $I_1$  and  $\Delta I$ .

In the record in figure 7(a), the pressure rose from 1 to 16.2 millimeters of mercury as the shock wave passed. The sharp initial rise of intensity is to about 0.10 inch on the record and the final drop after the mixing zone was to about 0.025 inch below the initial level. The ratio,  $I_2/I_1$  is then 5 to 1. It is seen that, when the glowing gas is suddenly compressed so that the concentrations of both the active and the normal nitrogen are changed simultaneously, the initial rise in the intensity of the afterglow is not proportional to pressure.

The intensity ratio may also be compared with the density ratio, the temperature ratio, and the ratio of the rate of bimolecular collisions ahead of and behind the shock. These ratios of temperatures, densities, and collision rates are related to pressures as described in appendix D. The pressure ratio of 16.2 to 1 corresponds to a density ratio of 4.4 to 1, a temperature ratio of 3.9, and a value of 8.5 for the ratio of the collision rates. The initial rise of intensity to 5 times the intensity before compression agrees roughly with the density ratio but not with collision rate, temperature, or pressure. These comparisons have a significant bearing on a choice between the different theories of the afterglow.

In evaluating the records of afterglow intensity, it should be pointed out that the photocell has a response that increases rapidly and in a nonlinear manner as the wavelength of the incident light decreases from the red to the blue. The ratio of 5 to 1 in the initial rise of the afterglow is acceptable then only if the spectral distribution does not change.

Effects of shock wave in different bands of afterglow.- Notable differences in response of the afterglow to shock waves were found when different optical filters were placed in front of the photocells. Figure 9 includes simultaneous records at four different pressures of the output from two photocells located at the same station of the shock tube. One photocell was masked by a filter transmitting the yellow band at 5750 angstrom units having a transmission band 80 angstrom units wide at half maximum. The other photocell was masked by a green filter having peak transmission at 5460 angstrom units with a band width of 200 angstrom units at half maximum. Both filters are of the interference type.

It will be noted in all four records that both yellow and green bands rise sharply at first and respond simultaneously to the arrival of the shock. The green reaches a maximum and then begins to decay while the yellow is still rising steadily. After about 1/2 millisecond, the yellow reaches a peak and begins to decay rapidly along with the green as the mixing zone passes the photocells.

The response of the afterglow in the green and yellow bands were then examined separately, using one channel of the oscilloscope for recording the intensity and the other channel for pressure.

Figure 10 shows a series of pressure and intensity records with a yellow filter in front of the photocell. It is seen that the initial rise of intensity is abrupt and that this is followed by a more gradual rise after the shock wave.

It is possible that the initial rise is proportional to the rise in density and that an additional slower effect associated with pressure is superposed. A rough separation of these effects was attempted on the records in figure 9, but it proved to be difficult to determine the magnitude of the initial rise because of the superposed effect of the slower and greater rise that followed. A further difficulty is encountered because of the possibility that the spectral distribution within the band passed by the filter is not constant and the response of the photocell varies greatly with wavelength in that region.

Figure 11 is a series of records of pressure and intensity taken with the green filter in front of the photocell. In each record, the intensity rises to a plateau that is well enough defined to allow approximate measurements of the magnitude for correlation with the

pressure records. The results are tabulated in table II. The static pressure in the shock tube  $p_1$  ahead of the shock is recorded from observation at the time of the test run. The pressure,  $p_1$ , was measured with a diaphragm type of absolute pressure gage. The readings are believed to be reliable to about 0.05 millimeter of mercury. The column  $\Delta p$  lists the pressure rise to the plateau occurring within 1/2 millisecond after the shock wave passed. The intensity  $I_1$  of the afterglow ahead of the shock is the deflection in inches of the trace on the record of the output of the photocell. The value of  $I_1$  is the difference between the deflection ahead of the shock and after the mixing zone has passed and the afterglow is extinguished. The value of  $\Delta I$  is the rise in inches from the initial level to the plateau on the photocell trace. The quasi-steady-state intensity after the shock  $I_2$  is the sum of  $I_1$  and  $\Delta I$ . The ratio of the density of the nitrogen ahead of and after the shock is determined from the pressure ratio  $p_2/p_1$  by use of the Rankine-Hugonot equations. Reference 17 includes a convenient tabulation of the values which were used in plotting the variation of  $\rho_2/\rho_1$  with  $P_2/P_1$  in figure 12.

A comparison of the values of  $\rho_2/\rho_1$  with  $I_2/I_1$  in table II shows strikingly good agreement. The evidence is strong that the intensity rise in the green band due to an abrupt adiabatic compression is proportional to the increase in density.

This result offers the possibility that the applications of the afterglow to the study of flow at low density may be extended to quantitative measurements of density. The result is significant also in its indication that energy is transferred to the levels from which afterglow is emitted at a rate that is proportional to density rather than collision frequency or temperature.

#### DISCUSSION OF RESULTS

Ionization. - The space discharges in the stream, apparently caused by free electrons moving with the active nitrogen, could be deflected strongly by magnetic fields. In contrast, the glowing stream of active nitrogen showed no response to magnetic fields transverse to the stream. The Stern-Gerlach experiment performed by Jackson and Broadway to measure the magnetic moment of active nitrogen (reference 14) also reported no deflection of the jet. If the jet of active nitrogen in their experiment had been composed of molecular ions as proposed by Mitra, a deflection of the jet should have been observed in addition to the splitting due to the molecular magnetic moment. The evidence, then, favors the view that ionization effects in the active nitrogen are the results of free electrons rather than positive ions in the stream.

The free electron density measured in the stream appears to be high enough to account for the glow discharges in the stream at regions of low pressure. In support of this view, it may be mentioned that active nitrogen is considered to be excited to states well above the ground state and the electric field required to cause a glow discharge is less in active nitrogen than normal nitrogen for that reason. It is presumed, then, that the active nitrogen breaks down into a glow discharge in those regions of expanding flow where the electric field, due to the streaming electrons, becomes sufficiently large.

Observations in the present tests agreed with those of previous tests by Rayleigh and others that the removal of ions from active nitrogen had no effect on the intensity or spectral distribution of the afterglow. The questions raised by Mitra regarding the use of probes in the earlier work to measure ionization were circumvented in the present tests by the use of a resonant cavity to determine that the ion trap was effective in reducing substantially the free electron density in the stream. The present results support the views of Gaydon, Constantinides (reference 18), and others that ionization in a stream of active nitrogen is incidental rather than essential to the production of afterglow.

Population of active centers.- The present measurements of heat released during decay of the afterglow indicated a population of active centers about  $1/6$  the maximum that Rayleigh measured by chemical methods. The maximum intensity of the afterglow per unit mass of nitrogen in the present tests was found to be about one-half the intensity obtained by Rayleigh. These comparisons indicate that greater excitation may increase the intensity of the afterglow in a stream by a factor of from 2 to 6 times that of the present tests.

The calorimeter measurements indicated that the population of active centers was approximately one in 250. This is of the same order as Rayleigh's chemical measurements of the population but is very much less than the values he obtained from the heating of metal sheets that he exposed to the active nitrogen.

Variation of intensity with pressure.- When the active nitrogen was suddenly compressed by a shock wave there was a sharp rise in the yellow part of the spectrum as well as the green. The initial rise in the intensity of both the yellow and green appeared to be proportional to the rise in the density. The later rise in the intensity of the yellow was a function of both the rise in pressure and the time after the sudden compression.

Delayed rise of yellow intensity.- The rise of the yellow intensity after a shock wave was found to continue almost linearly with time over a period of the order of a millisecond which was about as long as the process could be observed in the shock tube that was used. It may be suggested that an intermediate molecular state is involved in the transition from long-lived metastable states to the stable B levels and that the increased pressure favors a growth in population of those levels that lead to emission of the yellow. The interpretation of this selective intensification of the yellow band is open to question until more detailed information becomes available with better spectral resolution. This selective growth could be caused by the effects of pressure in perturbing adjacent potential curves, favoring transitions to some vibrational levels more than others.

Applications of nitrogen afterglow to low-density flow.- The preliminary investigation of nitrogen afterglow in visualizing low-density supersonic flow, reference 1, demonstrated that sharply defined changes in intensity coincided with shock waves. Uncertainties as to the processes causing the afterglow raised questions regarding the possibility of quantitative interpretations of the flow patterns to obtain measurements of density or pressure changes. Further questions were raised as to the extent to which the aerodynamic properties of active nitrogen would depart from those of normal nitrogen and air.

The present results show that the intensity of the afterglow on either side of a shock in a stream tube is not a simple, linear function of pressure, density, or temperature. The effects vary with time after the gas is compressed and are different in different parts of the spectrum. This variation with time after compression should be considered in the interpretation of afterglow patterns in low-density flow. The positions of shock waves will be correctly indicated, but the continuous variation in intensity of afterglow downstream of a shock may not be taken as a reliable indication in density unless the afterglow is viewed through a suitable filter. There is a strong indication that in certain portions of the spectrum of the afterglow, particularly the green band, the changes in intensity are a linear function of changes in density within the stream tube. This property of the afterglow offers the possibility that the active nitrogen may be used for quantitative measurements in low-density flow where conventional methods are of little or no use. The present tests showed this for the available range of static pressures ahead of the shock from 0.6 millimeter up to 20 millimeters and for pressure ratios across the shock up to about 15 to 1. Further tests with improved accuracy and provisions for a wider range of pressures and shock strengths would be desirable to determine the range of pressures within which the quantitative interpretation is practical and the precision that may be expected in different parts of the spectrum.

The results of the viscosimeter and calorimeter tests show that the viscosity and temperature of streaming active nitrogen are not substantially disturbed by processes associated with the afterglow. The viscosity was found not to change within the experimental error of 1/2 of 1 percent. The temperature was measurably affected but by amounts that are believed to be acceptable in experimental investigations of low-density flow.

The unusually high density of free electrons in a stream of active nitrogen should be given some further consideration in applications of the afterglow to low-density flow. Although the viscous forces did not appear to be affected, it is possible that disturbing effects may be found under conditions differing from those in the present tests.

The temperature of an excited gas may not be determined in a simple manner if the various degrees of freedom are not in equilibrium. Bethe and Teller (reference 19) have treated this problem in detail by considering separately the active and inert degrees of freedom of a gas flowing through a strong shock. The active degrees of freedom, rotation and translation, reach equilibrium in the order of one or two collisions after the shock and, in doing so, establish a characteristic temperature of the flow that may be measured by a conventional stagnation thermometer. The inert degrees of freedom require many more collisions before equilibrium is attained. In the case of vibration, as many as 500,000 collisions may be required for nitrogen. Because of this lag, the vibrational energy levels may be classed as metastable.

For active nitrogen, the long lifetime of the afterglow is in itself an indication that inert degrees of freedom are excited and that equilibrium may become established only after many collisions. This process leads to a gradual rise in the temperature of the active degrees of freedom during the period in which active nitrogen releases energy. Part of the energy may be radiated outside the stream tubes and have no effect on the temperature. Some part of the inert energy may be expected to pass by inelastic collisions or by another process to the active degrees of freedom and thereby raise the temperature as the activity decays. The results of the calorimeter tests provide an estimate of the amount of the temperature rise that may be expected to occur in this way.

The calorimeter measurements indicated that a temperature rise of as much as 90° C may be expected to occur in a stream while the active nitrogen is returning to its normal state. It is reasonable to question whether additional energy remaining after the decay of the visible afterglow may not cause a further increase. This appears to be unlikely since the spectral distribution of the afterglow, including the infrared region, is believed not to change as it decays at constant temperature and pressure (see reference 13). The time during which the decay occurs is long compared to the time of 1 or 2 milliseconds required for active nitrogen to travel through a supersonic nozzle or other



aerodynamic test section of flow. If the half-life of the afterglow is 1 second, a temperature rise of about  $0.05^{\circ}\text{C}$  may be expected in a stream tube during a time of 0.001 second required for an element of the active gas to move a distance of the order of 2 feet at supersonic velocities. A temperature rise of this amount is considered to be acceptable in aerodynamic applications of the afterglow.

Suggestion for further research using shock tubes.- The results obtained in the study of active nitrogen by the use of a shock tube suggest that the methods described in appendix D are applicable to more detailed studies of shock waves at low density. The results demonstrate that traveling shock waves in active nitrogen may be observed, and wave velocity and probably density may be measured with usable precision. The possibilities are especially interesting in that the response of the photocells used in observing the effects is as fast as interferometric methods and a great deal faster than can be expected with pressure gages or hot-wire devices customarily used in this type of work at higher densities.

The response of the different bands in the afterglow, including the red and infrared bands, to compression may be studied in more detail by the use of shock tubes and photocells in much the same way as described herein. It appears to be feasible to use a monochromator having sufficient dispersion to resolve the vibrational structure of the bands with adequate transmission of light to operate the photocells. Another possibility for obtaining detailed information within the bands is to flow the active nitrogen through a nozzle designed to cause a stationary shock wave ahead of a relatively long section that is free of shock waves. This would allow the use of a conventional spectrograph, but the design of the nozzle with a suitable turning angle presents a difficult problem, especially at the low pressures that would be desirable.

This additional research, either by the use of shock tubes or nozzles, would be useful in arriving at a suitable explanation of the differences in response that were observed in the green and yellow bands. The differences may be found to be related in some way to the long lifetime of vibrational degrees of freedom. Deviations from thermal equilibrium in a shock wave are known to occur because of the lag in establishing equilibrium between the vibrational degree of freedom and the active degrees of freedom. Nitrogen is notable in that the lag is greater than that of most gases (table V and part 2(d) of reference 19).

This lag may be expected to play a part both in the shock waves and the afterglow.<sup>1</sup>

### CONCLUSIONS

The experimental investigation of active nitrogen in low-density flow, under the conditions prevailing in the experiments, resulted in the following conclusions:

1. In the use of active nitrogen for studies of low-density flow by observation of the afterglow, disturbances are introduced in the form of heating and ionization, but the disturbances appear not to be so large as to interfere with its use for this purpose.

2. The Lewis-Rayleigh afterglow in active nitrogen responds to the compression at a shock front with adequate speed to correctly define the position of the shock wave.

3. Following the initial rise in the intensity of the afterglow, there is a gradual rise downstream of a shock wave in certain portions of the spectrum. This delayed rise may prevent reliable interpretation of continuous changes in density of the flow downstream of a shock unless the afterglow is viewed through a filter passing only a portion of the spectrum, such as the green band of the afterglow.

4. Transient variations in the intensity of the afterglow may be used as a measure of variations of density in the stream in certain restricted regions of the spectrum.

5. Unless a suitable ion trap is provided, space discharges may occur in expanding regions of the flow because of free electrons moving with the stream.

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<sup>1</sup>Professor Joseph Kaplan first suggested to the writer that the heat capacity lag of nitrogen may be found to be an essential part of the phenomena associated with the Lewis-Rayleigh afterglow. His suggestion seems plausible and it is conceivable that long-lived vibrational levels of metastable states may undergo radiationless transitions to the  $B^3\Pi_g$  state, from which the afterglow is emitted. The vibrational levels of the  $a^1\Sigma$  and the  $w$  electronic states appear to have sufficient energy to take part in the process.

6. Ionization in a stream of active nitrogen is incidental rather than essential to the production of the afterglow.

7. The observed response of the afterglow to shock waves is evidence that active nitrogen includes nitrogen molecules in metastable levels adjoining the B state.

Langley Aeronautical Laboratory  
National Advisory Committee for Aeronautics  
Langley Field, Va., December 18, 1950

## APPENDIX A

## APPARATUS FOR PUMPING AND ACTIVATING THE STREAMING NITROGEN

The apparatus for activating the stream of nitrogen is illustrated in figure 13. The nitrogen flows from a standard commercial cylinder through a pressure-reduction valve and a flowmeter. A further reduction in pressure occurs across the needle valve before it enters the exciter tube. The flowmeter is of a conventional type manufactured by Fisher and Porter.

The exciter tube is made of Corning Vycor glass, 50 centimeters long, 25 millimeters outside diameter and with a wall about 1.5 millimeters thick. Three pairs of side arms 12 millimeters outside diameter and 8 centimeters long support the electrodes. The arms are spaced 13 centimeters apart. The electrodes were aluminum rods tipped with a tungsten wire about 1 millimeter in diameter which extends about 8 millimeters from the end of each aluminum rod. Each rod is held in place by a Neoprene stopper at the outer end and asbestos tape wrapped around the inner end to form a bushing that prevents the heated electrode from touching the glass tube.

Each of the three electrical discharges was powered by a General Electric Company transformer, type 51G302 rated at 825 V.A., at 15,000 volts output with an input of 115 volts at 60 cycles per second. This type is commonly used for operating neon signs. The secondary was connected across a condenser having a capacity of approximately 0.01 microfarad. In series with the exciting discharge was a spark gap of about 4 millimeters between tungsten electrodes 4.5 millimeters in diameter.

The flow of gas is maintained by the pumping system illustrated in figure 14 showing the supersonic nozzle in place.

The booster pump was used only when it was desired to pump at pressures in the range below 1 millimeter. The mechanical pump will maintain a slow rate of flow through the shock tube at a pressure of 0.5 millimeter, but it is necessary to use the booster pump to operate the nozzle at a static pressure of 0.5 millimeter and below in the supersonic region.

The nozzle was designed by the method of characteristics for a Mach number of 3.59 at pressures considerably higher than used in the present tests. It is generally known that nozzles designed by this method do not develop uniform flow at the design Mach number at low densities because of the effects of boundary layers that are difficult

to predict. Completely satisfactory methods of producing low-density flow in a nozzle have not yet become available and it was necessary to carry out the present tests with the available design. Because of the cross-stream gradients in the nozzle, it was considered of no significance to include in the present results any measurements of the change in brightness of the afterglow as the nitrogen expands through the nozzle to a pressure of about 1/20 the pressure in the upstream end of the nozzle.

The nozzle was used in observations of the space discharge in expanding flow and in obtaining the spectrum of the afterglow in the region where the pressure in the stream decreases rapidly.

The nitrogen used for all the experimental work was obtained from commercial sources and was of the grade designated "dry" nitrogen. The impurities in a typical sample were reported by the manufacturer to be:

Water vapor, percent by weight . . . . .	0.03
Hydrogen, parts per million . . . . .	Less than 20
Oxygen, percent by volume . . . . .	Not more than 0.3
Argon, percent . . . . .	0.06

## APPENDIX B

## VISCOSIMETER AND CALORIMETER

Measurement of viscosity.- The viscosity of the active nitrogen was compared with that of normal nitrogen in a viscosimeter arranged as in figure 15. The gas was flowed slowly through a fixed length of tubing at constant temperature. A butyl phthalate manometer measured the pressure drop due to the laminar flow through the tube. The pressure drop was observed with and without electrical excitation while the mass flow was held to a constant value.

Release of heat in active nitrogen.- The apparatus sketched in figure 15, with the addition of a resistance thermometer, was used as a calorimeter. The excited gas flowed slowly through the tube in the water bath and the temperature rise was observed for runs lasting as long as 2 hours. Check runs were made with the stream of nitrogen unexcited.

The tubing was of 2S aluminum alloy,  $\frac{3}{8}$  inch inside diameter, in the form of a helical coil having eight turns and a helix diameter of approximately  $4\frac{1}{2}$  inches. Another coil of copper having the same dimensions replaced the aluminum for a few runs to compare the release of heat by active nitrogen on copper with that released on aluminum.

A resistance thermometer of 99-percent nickel alloy was used to measure the temperature changes in the water surrounding the helical coil. The thermometer had a nominal resistance of 100 ohms and a sensitivity of approximately 0.43 ohms per degree centigrade. Resistance readings were readily made to an accuracy of 0.005 ohm corresponding to an accuracy in reading temperature changes of  $\pm 0.012^{\circ}$  C.

At the start of a calorimeter run, the water bath was at practically room temperature. Readings were taken at intervals during a run of the order of 1 hour in duration. The vacuum flask and surrounding insulation of cotton prevented any appreciable loss of heat from the water bath during the run. This was established by observing that the temperature of the water remained constant within the accuracy of the measurement for an hour or more after the end of a run.

Two thermocouples of iron constantan were placed in the stream of active nitrogen to measure the drop in temperature of the gas in flowing through the calorimeter. This "temperature" indicated by the thermocouple is considered to be that characteristic of the active degrees of freedom which are translation and rotation. The effects of vibrational

and possibly electronic degrees of freedom of long lifetime may be involved in the production of afterglow and are detected by the heating effects that accompany the decay of the afterglow in the calorimeter. In this fashion, the heat transferred to the calorimeter during a run is considered as made up of one part due to the drop in temperature shown by the thermocouples and another part due to the conversion of inactive forms of energy into heat by some process that is responsible also for the visible afterglow. It is the latter part of the heat that is taken to be indicative of the energy carried in latent form by the active nitrogen.

Provision was made for making test runs with the calorimeter coil either grounded or insulated from ground to observe the heating effects of ions striking the coil in passing from the stream to ground.

Consideration was given to the possibility that active nitrogen may affect the thermocouple readings in the way that metal foils were heated in Rayleigh's experiments. An examination of the data for any of the calorimeter tests will show that the heat flow to the calorimeter, as indicated by the two thermocouple readings, was in every case much less than the total heat flow. The thermocouple at the exhaust end always read about the same as the temperature of the water in the calorimeter. This demonstrated that the thermocouple in the incoming active nitrogen was not greatly heated by the release of latent energy from the stream.

Mass flow of the gas was measured by means of Fisher and Porter "flowrator" No. J7-1432 having a useful range from 100 to 1000 cubic centimeters per minute at 14.7 pounds per square inch pressure. The present tests were made with a pressure of 40 pounds per square inch and a temperature of about 75° F in the flowmeter and with a volumetric flow of approximately 975 cubic centimeters per minute. The mass flow was computed from readings of the flowmeter by the procedure outlined in the manufacturer's instruction sheets (catalog section 98-A (1947), Fisher and Porter Company, Hatboro, Pa.).

## APPENDIX C

## MEASUREMENT OF ELECTRON DENSITY BY MICROWAVE METHODS

The symbols used in this appendix are as follows:

$\lambda$	resonant wavelength of cavity with electrons in stream
$\lambda_0$	resonant wavelength of cavity without free electrons in the stream
$\sigma_i$	imaginary part of conductivity of the gas
$\sigma_r$	real part of conductivity of the gas
$E$	electric field in the cavity
$c$	velocity of light
$\epsilon_0$	permutivity of free space
$n$	free electron density
$e$	electronic charge
$m$	mass of the electron
$E_r, E_\theta, E_z$	components of electric field along respective axes of cylindrical coordinates
$J_0, J_1$	Bessel functions of the first kind, zeroeth and first order

The methods described in reference 20 were applied to the measurement of free electron density in the active nitrogen. The apparatus is illustrated in figure 16. The active nitrogen flows through a Vycor tube 10 millimeters inside diameter and 12 millimeters outside diameter that is coaxial with a cylindrical cavity 76.5 millimeters inside diameter and 109 millimeters long. The cylinder has a resonant wavelength of about 10 centimeters when oscillating in the  $TM_{010}$  mode.

The cavity is made of steel with the inner surface silver-plated to reduce losses in the skin. Two magnetic loops are provided, one to drive the cavity and one to lead a signal out to the receiver. The cables leading from the generator are each 12 feet long and of AN type RG 21/U having an attenuation of 0.82 decibel per foot at 3000 megacycles.



This attenuation was provided to avoid excessive loading and frequency shift of the klystron generator.

The resonant frequency of the cavity was measured with the vycor tube in place but without active nitrogen streaming through. The resonant frequency was then measured with active nitrogen flowing through the cavity. The shift in resonant frequency due to the complex conductivity of the gas is a measure of the free electron density in the stream.

The change in resonant wavelength is given by (reference 21)

$$\frac{\Delta\lambda}{\lambda} = - \frac{\lambda_0 \int_{V_1} \sigma_i E^2 dv}{4\pi c \epsilon_0 \int_{V_2} E^2 dv} \quad (1)$$

provided that

$$\sigma_i \ll \epsilon_0 \frac{2\pi c}{\lambda_0}$$

and

$$\sigma_r \ll \sigma_i$$

The electron density and  $\sigma_i$  are related by

$$\sigma_i = - \frac{ne^2\lambda}{2\pi mc} \quad (2)$$

The contribution to  $\sigma_i$  of any positive ions that may be present in concentrations comparable to that of electrons is negligible because of the large mass of the ions. In the present case, the collision frequency between electrons and molecules is small compared to the frequency of the microwaves and

$$\sigma_r \ll \sigma_i$$

Equation (1) may be solved for  $\sigma_i$  by evaluating the two definite integrals. The electron density is assumed to be constant throughout the stream and the electric field is integrated over the two volumes, that enclosed within the vycor tube and that extending throughout the cavity. It is sufficient for the present purpose to neglect the

distortion in the field due to the glass tube and write for the electric field in a  $TM_{010}$  cavity:

$$E_r = E_\theta = 0$$

$$E_z = E_0 J_0 \left( \frac{2.4}{R} r \right)$$

Equation (1) becomes on substitution of  $E_z$  for  $E$  and of  $r d\theta dr dz$  for  $dv$

$$\begin{aligned} \frac{\Delta\lambda}{\lambda} &= - \frac{\lambda_0 \sigma_i E_0^2 \int_0^{2\pi} \int_0^a \int_0^h r \left[ J_0 \left( \frac{2.4}{R} r \right) \right]^2 d\theta dr dz}{4\pi c \epsilon_0 \int_0^{2\pi} \int_0^R \int_0^h r \left[ J_0 \left( \frac{2.4}{R} r \right) \right]^2 d\theta dr dz} \\ &= - \frac{\lambda_0 \sigma_i E_0^2 \int_0^a r \left[ J_0 \left( \frac{2.4}{R} r \right) \right]^2 dr}{4\pi c \epsilon_0 \int_0^R r \left[ J_0 \left( \frac{2.4}{R} r \right) \right]^2 dr} \\ &= - \frac{\lambda_0 \sigma_i E_0^2 \frac{a^2}{2} \left\{ \left[ J_0 \left( 2.4 \frac{a}{R} \right) \right]^2 + \left[ J_1 \left( 2.4 \frac{a}{R} \right) \right]^2 \right\}}{4\pi c \epsilon_0 \frac{R^2}{2} \left\{ \left[ J_0(2.4) \right]^2 + \left[ J_1(2.4) \right]^2 \right\}} \\ &= - \frac{\lambda_0 \sigma_i E_0^2 \frac{a^2}{R^2} \frac{J_0^2 \left( 2.4 \frac{a}{R} \right) + J_1^2 \left( 2.4 \frac{a}{R} \right)}{0.269}}{4\pi c \epsilon_0} \end{aligned}$$

For the present apparatus

$$\frac{a}{R} = \frac{0.2}{1.53}$$

and

$$\frac{\Delta\lambda}{\lambda} = - \frac{\lambda_0 \sigma_1 E_0^2}{4\pi c \epsilon_0} \left( \frac{0.2}{1.53} \right)^2 0.975$$

Substituting for  $\sigma_1$  and replacing  $\lambda_0$  by  $\lambda$ , which is permissible, since

$$\Delta\lambda \ll \lambda$$

gives

$$\frac{\Delta\lambda}{\lambda} = \frac{\lambda^2 E_0^2}{4\pi c \epsilon_0} \frac{ne^2}{2\pi mc} \left( \frac{0.2}{1.53} \right)^2 0.975$$

Substituting numerical values in this equation yields

$$n = \frac{\Delta f}{f} (2.73 \times 10^{12}) \text{ electrons/cc}$$

or  $n = 1.05 \times 10^9$  electrons per cubic centimeter for 1-megacycle shift in resonant frequency of the cavity.

## APPENDIX D

## THE SHOCK TUBE

The shock tube with associated instruments was arranged as illustrated in figure 17 to measure changes in brightness and pressure when strong shock waves are passed through active nitrogen. The tube is of pyrex glass 50 millimeters in diameter and 12 feet long from the exciter tube to the side arm leading to the vacuum pumps. The pressure gage is of the "subminiature" variable-inductance type developed at Langley Aeronautical Laboratory. The photocells are RCA 931-A multiplier cells with maximum sensitivity in the blue. The oscilloscope is a DuMont type 279 having two beams.

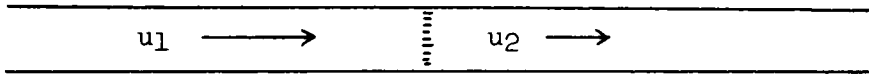
A steady-state flow of active nitrogen at low speeds was first established in the glass shock tube and the static pressure adjusted by controlling the mass rate of flow. The gains in the photocell and pressure circuits were adjusted to give desired amplitudes on the oscilloscope when the shock wave passed. For each run, the camera was started and allowed to accelerate to near maximum speed and the diaphragm was punctured. The inflow of atmospheric air started a shock wave down the glass tube and also into the line leading to the pump. The shock passed the photocell causing a rise in brightness followed by a decay when the mixing layer between active nitrogen and air passed.

A cathode follower was connected by short leads to the photocell, allowing the use of cables up to 100 feet without excessive lag in response. Much of the recording apparatus was removed to a distance where it was not subject to strong interference from the exciter discharge.

The mixing region between the active nitrogen and the incoming air registers as a darkening region as it passes the photocell. This region, which is often referred to as the cold front in a shock tube, follows the shock wave, but at a lower velocity. The velocity of this region may be obtained from the interval between the decay of the glow at two different stations. The characteristics of flow in a shock tube are well described in reference 16. Numerical values of pressure, density, and temperature changes across shock waves normal to the flow are tabulated for air in reference 17. The values for air were considered sufficiently close to those for nitrogen for use in the present work.

The compressible flow tables of reference 17 list the Mach number for the flow on either side of the shock. The Mach number, temperature, and collision frequency of the molecules before and after a shock wave are shown to be related as follows:

In the stream tube illustrated below, the flow is from left



to right through the stationary shock wave. Continuity of mass requires

$$\frac{u_1}{u_2} = \frac{\rho_2}{\rho_1}$$

Also,

$$M_1 = \frac{u_1}{a_1} = \frac{u_1}{\sqrt{\gamma RT_1}}$$

$$\theta_1 = KP_1\sqrt{T_1}$$

$$\frac{M_1}{M_2} = \frac{\rho_2}{\rho_1} \frac{\sqrt{T_2}}{\sqrt{T_1}} = \frac{\theta_2}{\theta_1}$$

$$\frac{T_2}{T_1} = \left( \frac{\rho_1 M_1}{\rho_2 M_2} \right)^2$$

where

- u      velocity of stream
- p      pressure
- $\rho$      density
- a      velocity of sound
- T      absolute temperature
- M      Mach number
- $\theta$      collision frequency

- K a constant involving collision cross section
- $\gamma$  ratio of specific heats
- R gas constant

The pressure gage is of the variable-inductance type differing from conventional design because of its small size. It incorporates a flat, stretched diaphragm of an alloy of iron containing 42-percent nickel having a thickness of 0.001 inch. The active area of the diaphragm is circular and has a diameter of 0.12 inch. For the present tests, the air gap was set at 0.0015 inch. The mechanical natural frequency of the gage is approximately 30 kilocycles per second.

The carrier circuit and amplifier used with the gage are of conventional design. The carrier frequency is 25 kilocycles per second. The over-all response of the electrical circuit and alternating-current bridge at the output was found to be flat within experimental accuracy up to 5000 kilocycles per second. Filtering circuits caused the response to fall off rapidly at higher frequencies.

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TABLE I.- MICROWAVE MEASUREMENTS  
IN ACTIVE NITROGEN

Run	Pressure (mm Hg)	Frequency, unexcited, (mc)	Frequency, excited, (mc)	Frequency shift (mc)	Remarks
1	0.2	2601.27	2601.37	----	Grounded
2	.2	1.13	1.18	----	Ungrounded
3	1.0	1.13	1.21	----	Grounded
4	1.0	1.12	1.22	----	Ungrounded
5	4.0	1.21	1.30	----	Grounded
6	4.0	1.21	1.12	----	Ungrounded
7	8.0	1.18	1.23	----	Grounded
8	8	1.00	1.34	0.34	Ungrounded
9	12	1.00	1.74	.74	Grounded
10	12	1.20	2.08	.88	Ungrounded
11	20	0.91	1.57	.66	Grounded
12	20	.96	1.47	.51	Ungrounded
<sup>a</sup> 13	12	1.10	2.08	.98	No ground
<sup>b</sup> 14	12	1.08	2.92	1.84	-----

<sup>a</sup>Aluminum grounding tube replaced by glass for run 13

<sup>b</sup>Shortened tubing from 3 feet to 1 foot of aluminum tubing between exciter and resonant cavity for run 14. Tubing grounded.



TABLE II.- DATA FROM RECORDS OF PRESSURE AND INTENSITY  
IN FIGURE 11 FOR THE GREEN BAND

Figure	$P_1$ (mm)	$\Delta p$ (mm)	$P_2/P_1$	$\rho_2/\rho_1$	$I_1$ (in.)	$\Delta I$ (in.)	$I_2/I_1$
11(a)	0.6	5	9.3	3.7	0.16	0.45	3.8
11(b)	1.0	9.6	10.6	3.9	.15	.42	3.8
11(c)	2.0	13.0	7.5	3.4	.14	.40	3.2
11(d)	5.0	27.0	6.4	3.2	.15	.35	3.3
11(e)	10	34	4.4	2.6	.16	.27	2.7

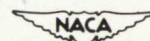
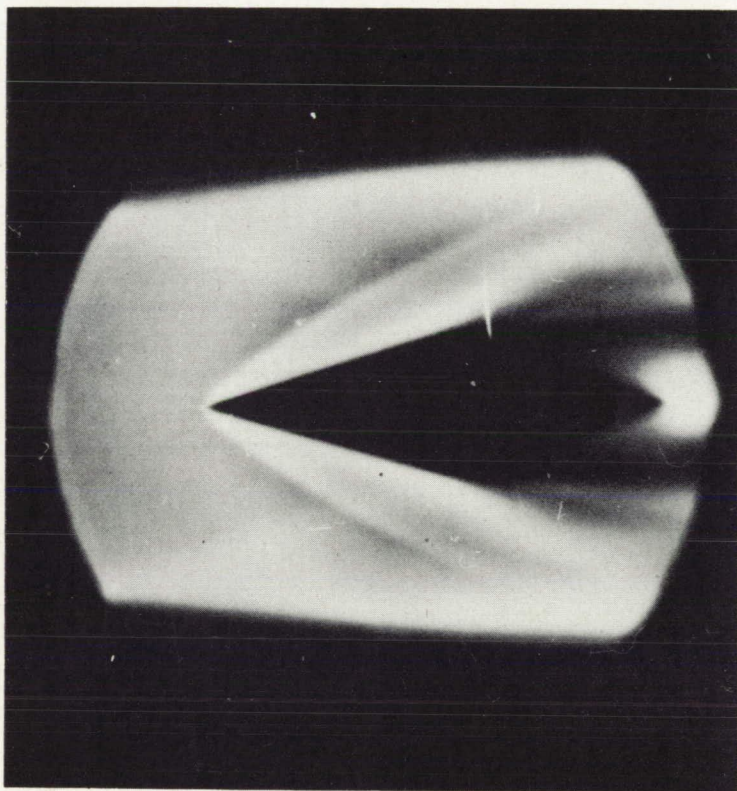




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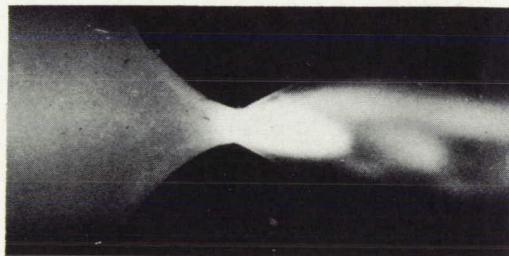


L-67967

Figure 2.- Photograph of flow over a double wedge showing a corona discharge in the low-pressure region around the trailing edge. Mach number, 2.6.



(a) Ions removed by grounded conducting surface exposed to the stream.



(b) Ground disconnected from conducting surface.



L-67968

Figure 3.- Photograph of separated flow through a supersonic nozzle with and without ion trap in the stream.

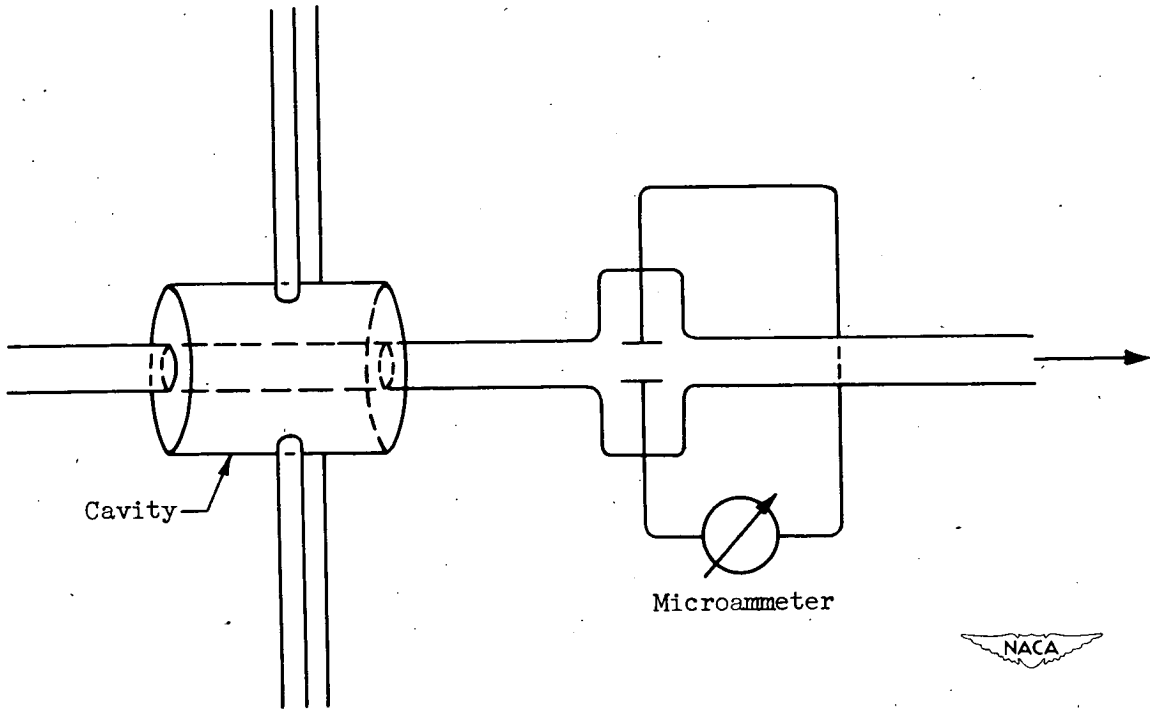
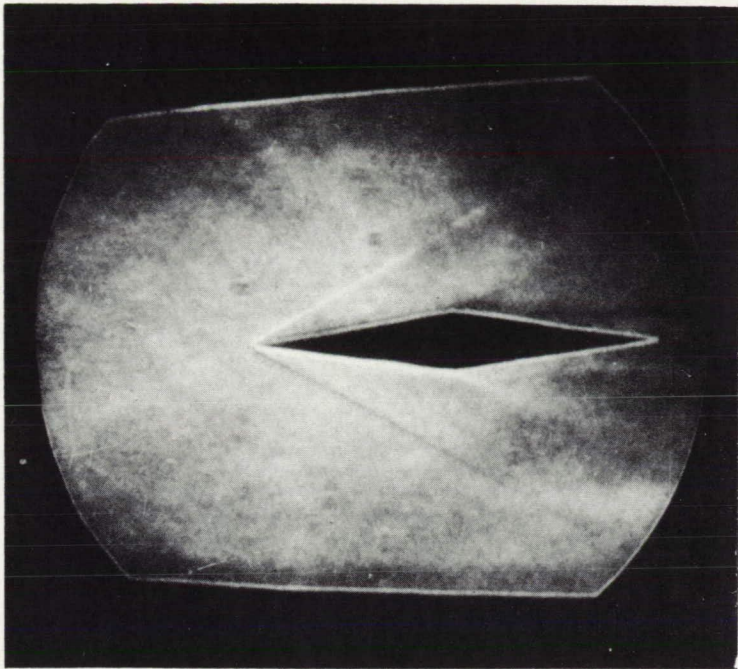
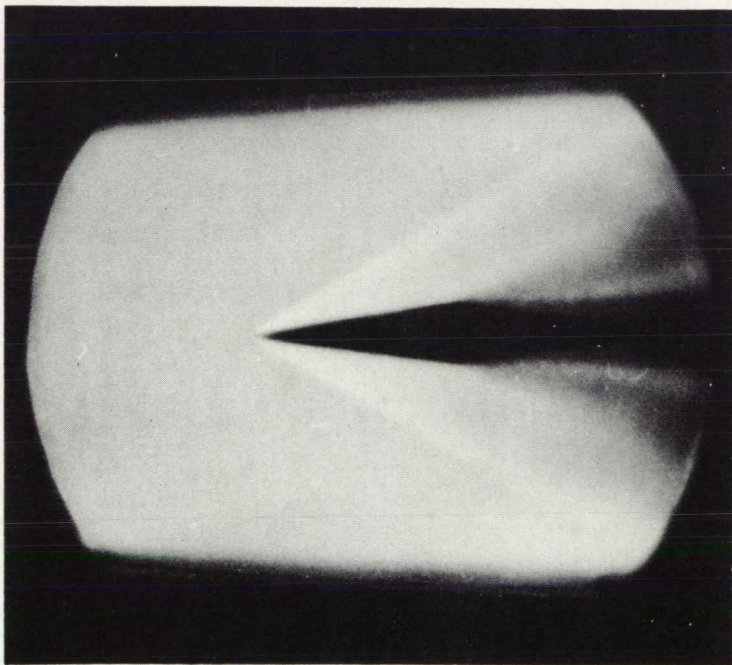


Figure 4.- Sketch showing location of electrodes in stream of active nitrogen near the resonant cavity.





(a) Schlieren.



(b) Afterglow.



L-67969

Figure 5.- Schlieren and afterglow photographs of supersonic flow over a double wedge. Mach number approximately 2.6. (From reference 1.)



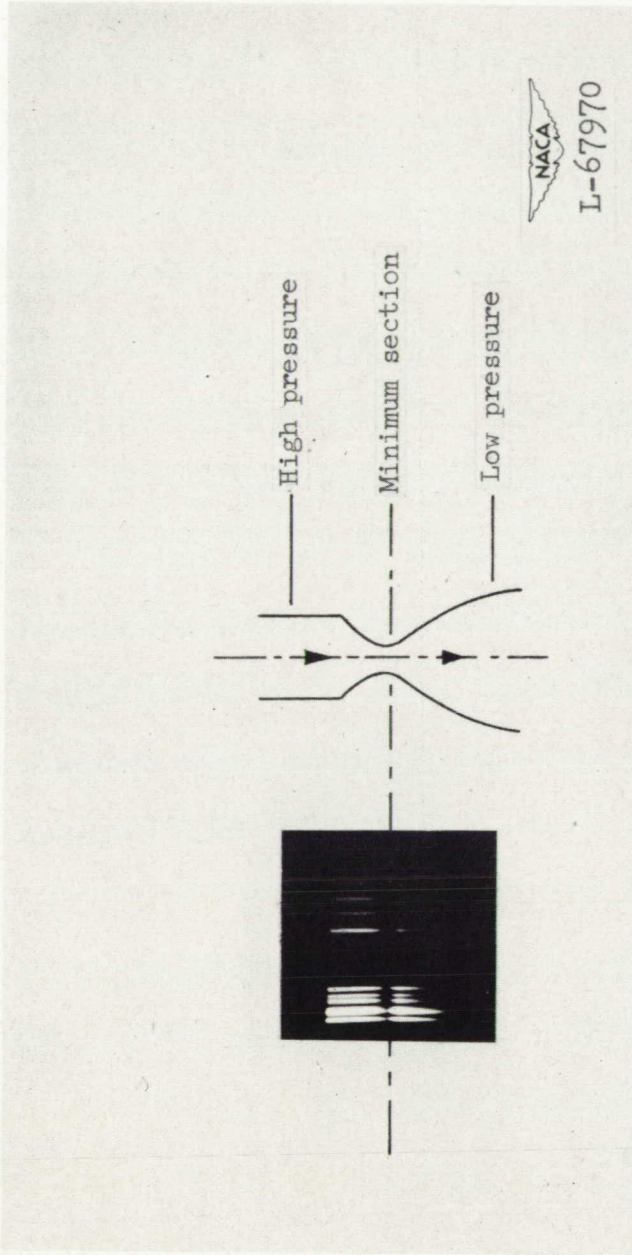


Figure 6.- Spectrum of afterglow in expanding flow at minimum section of nozzle. Upper end of slit exposed to settling chamber for 2 hours. Lower end exposed 4 hours to expanding flow in supersonic portion of nozzle. The break in the spectrum lines is at the minimum section.

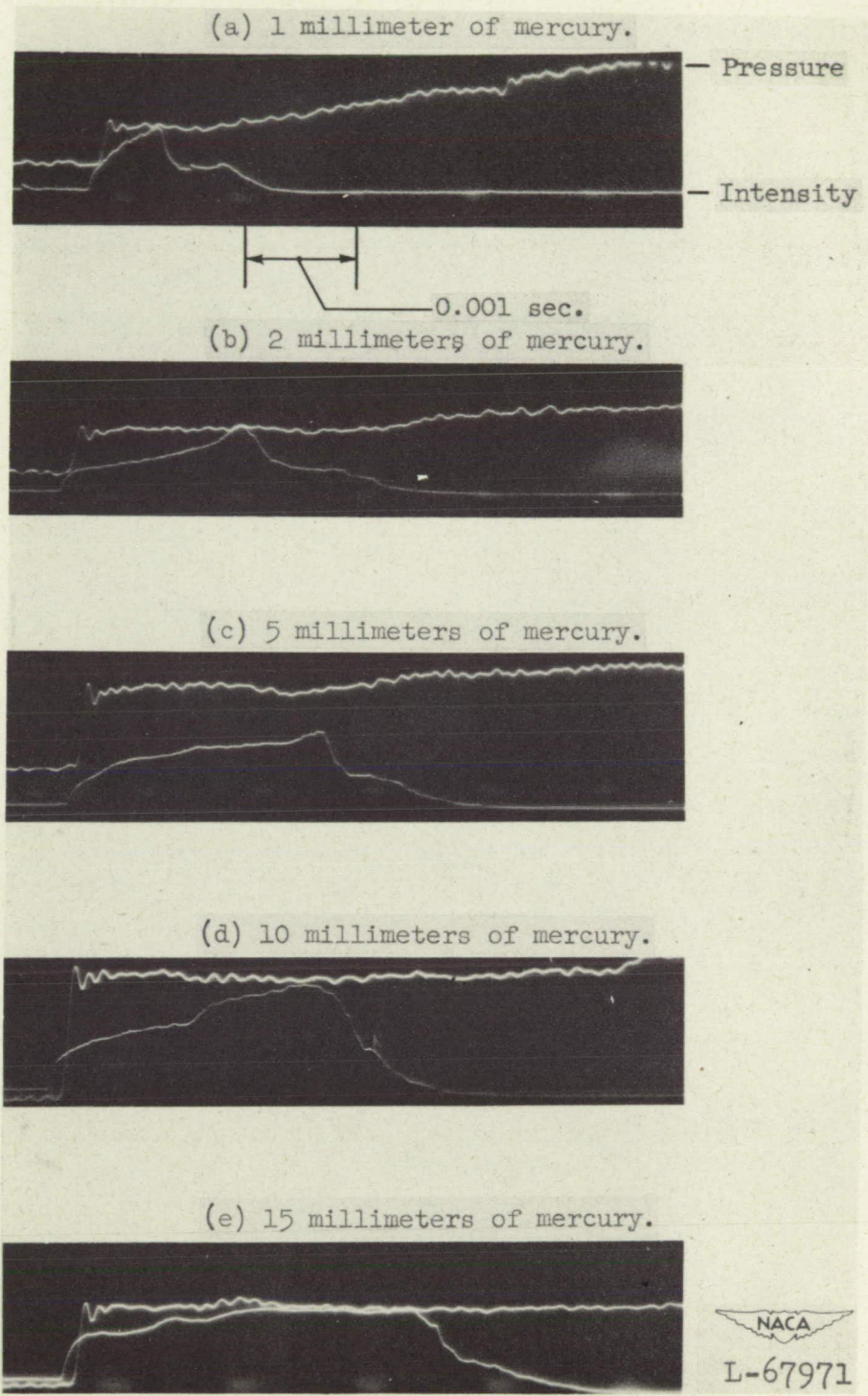
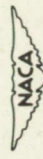
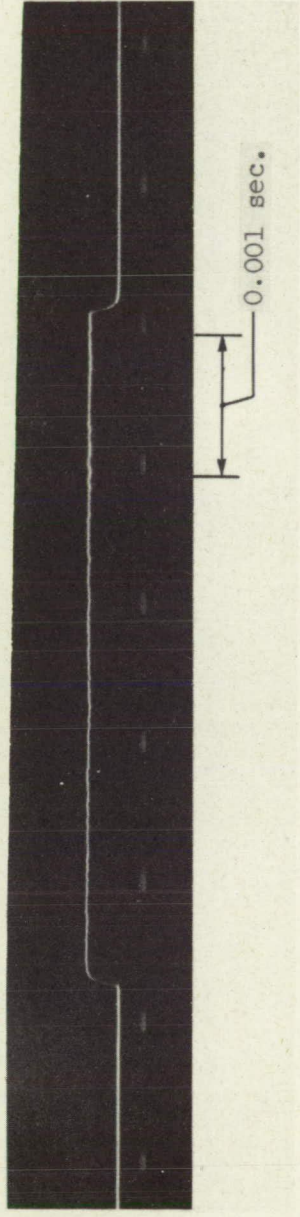


Figure 7.- Records of variation of pressure and intensity of afterglow across a shock wave at five different pressures.





L-67972

Figure 8.- Record of photo cell response to light pulsed by a chopper.  
Timer marks are spaced 1 millisecond apart.

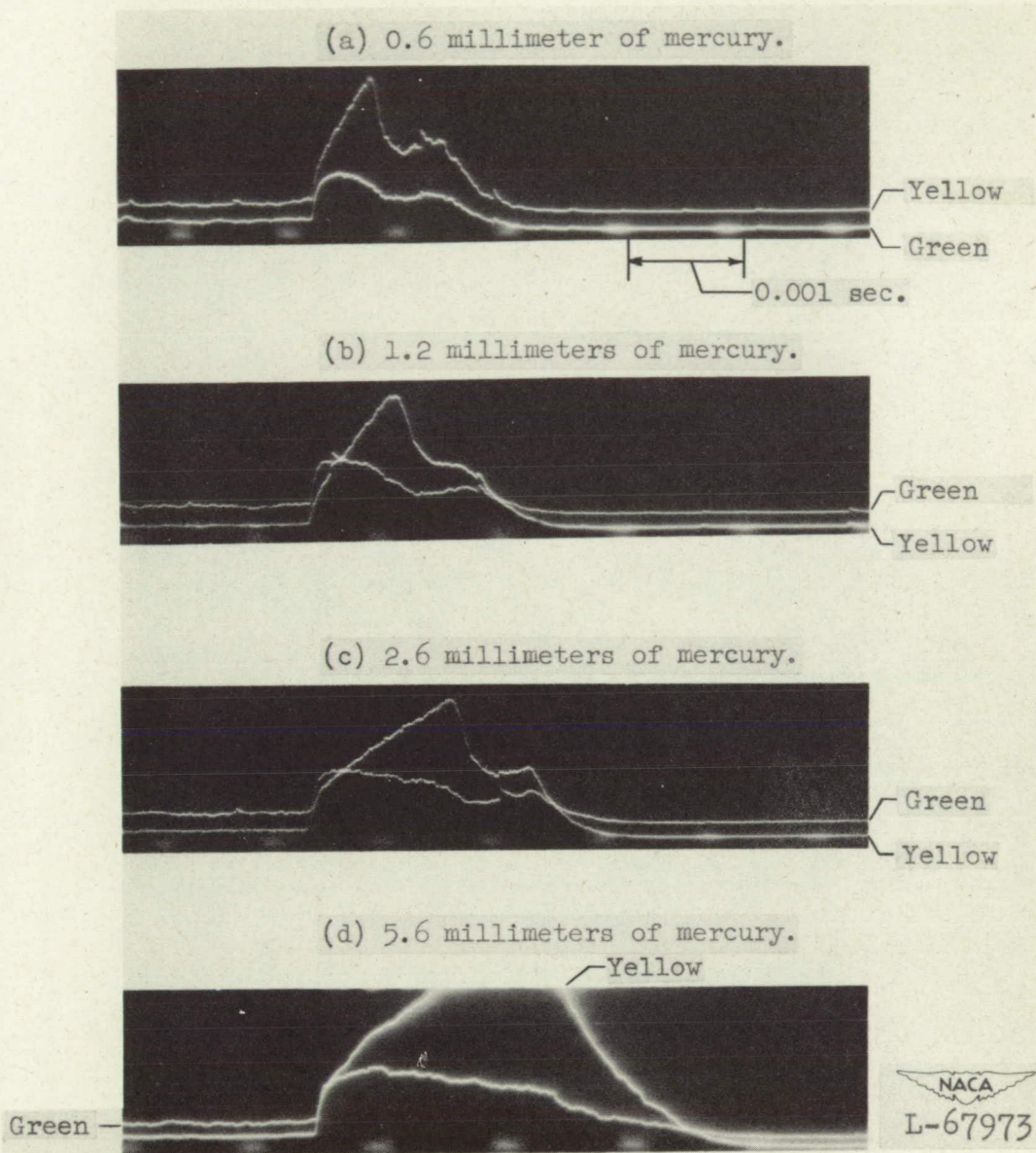


Figure 9.- Records of intensity changes across shock waves in afterglow at four different initial pressures. A green interference filter having peak transmission at 5460 angstrom units was in front of one photocell. A yellow interference filter having a peak transmission at 5750 angstrom units was in front of the other photocell.



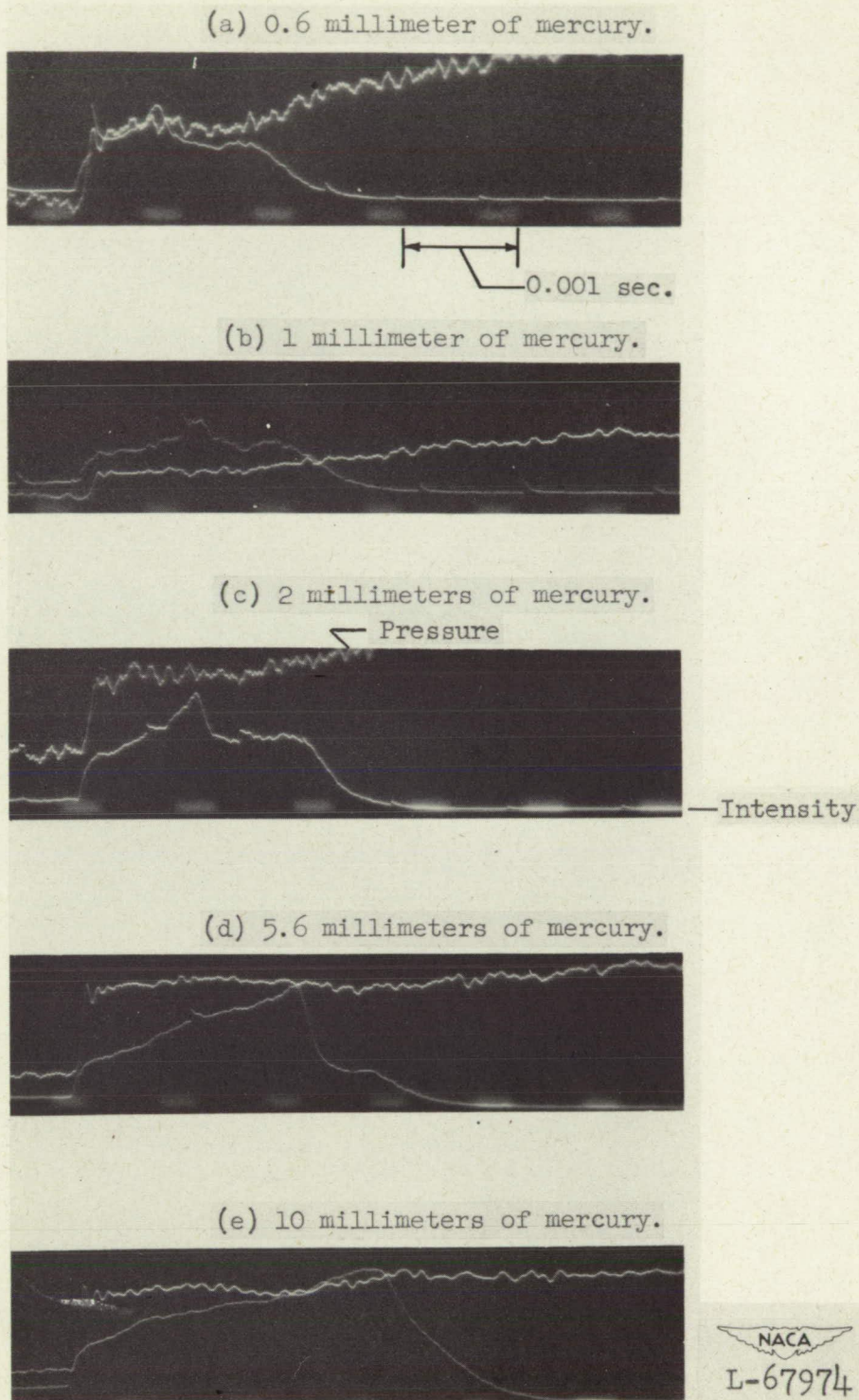


Figure 10.- Records of pressure and intensity changes in the yellow band across a shock wave in active nitrogen, for different static pressures ahead of the shock.

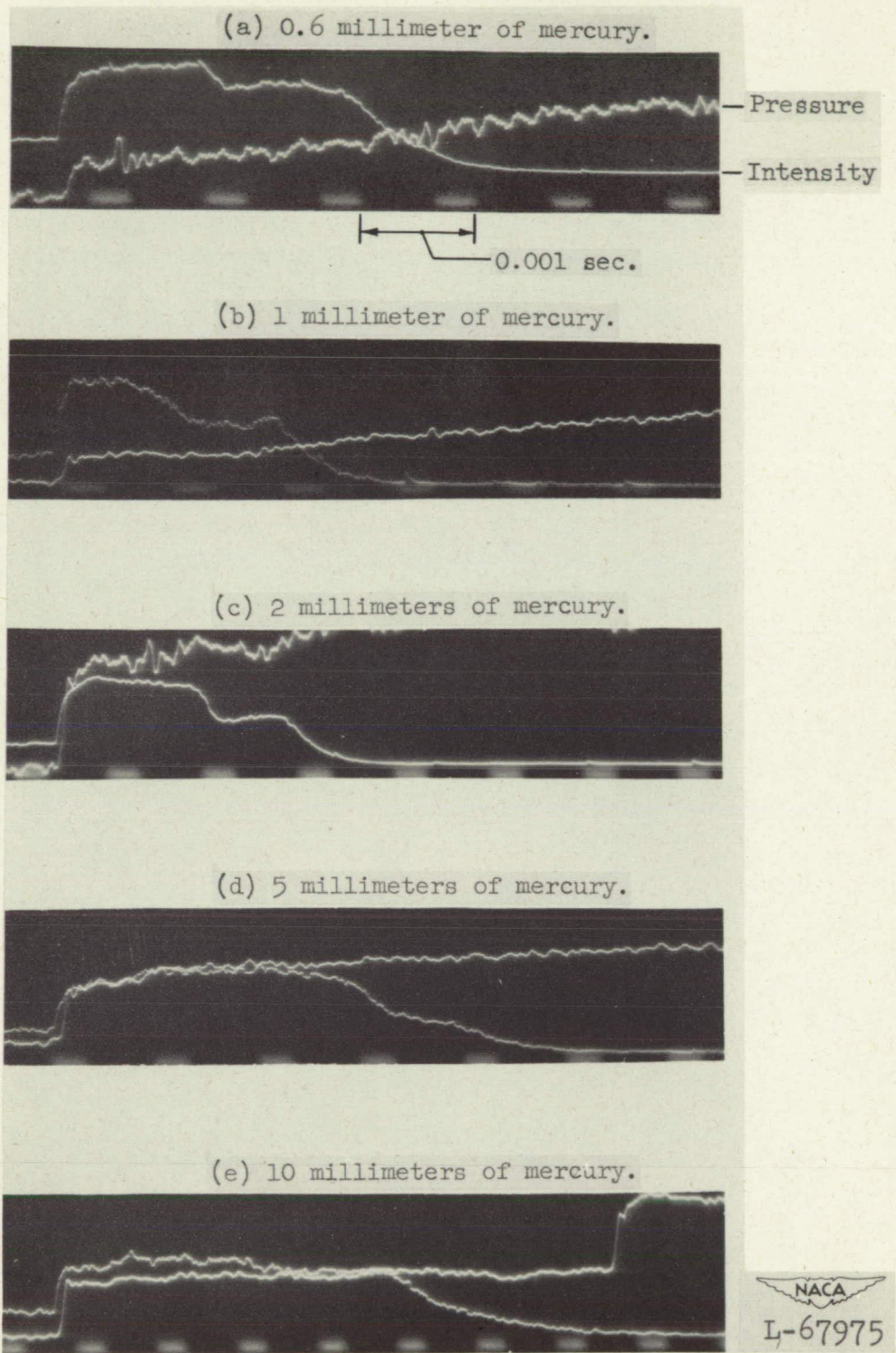


Figure 11.- Records of pressure and intensity changes in the green band across a shock wave in active nitrogen, for different static pressures ahead of the shock.



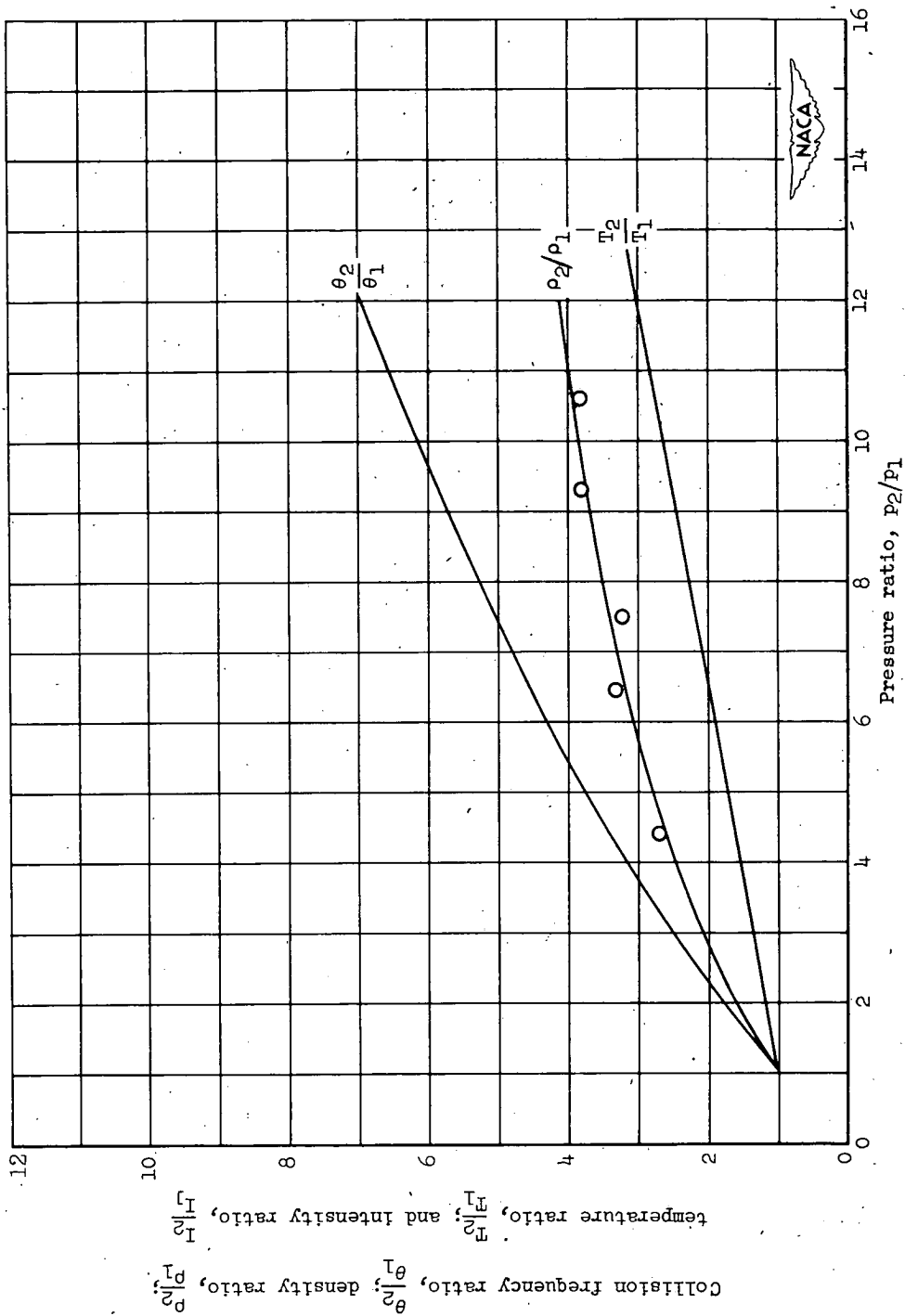


Figure 12.- Calculated variation of density ratio, collision frequency ratio, and temperature ratio with pressure ratio across a normal shock. Plotted points are intensity ratios measured in the green band.

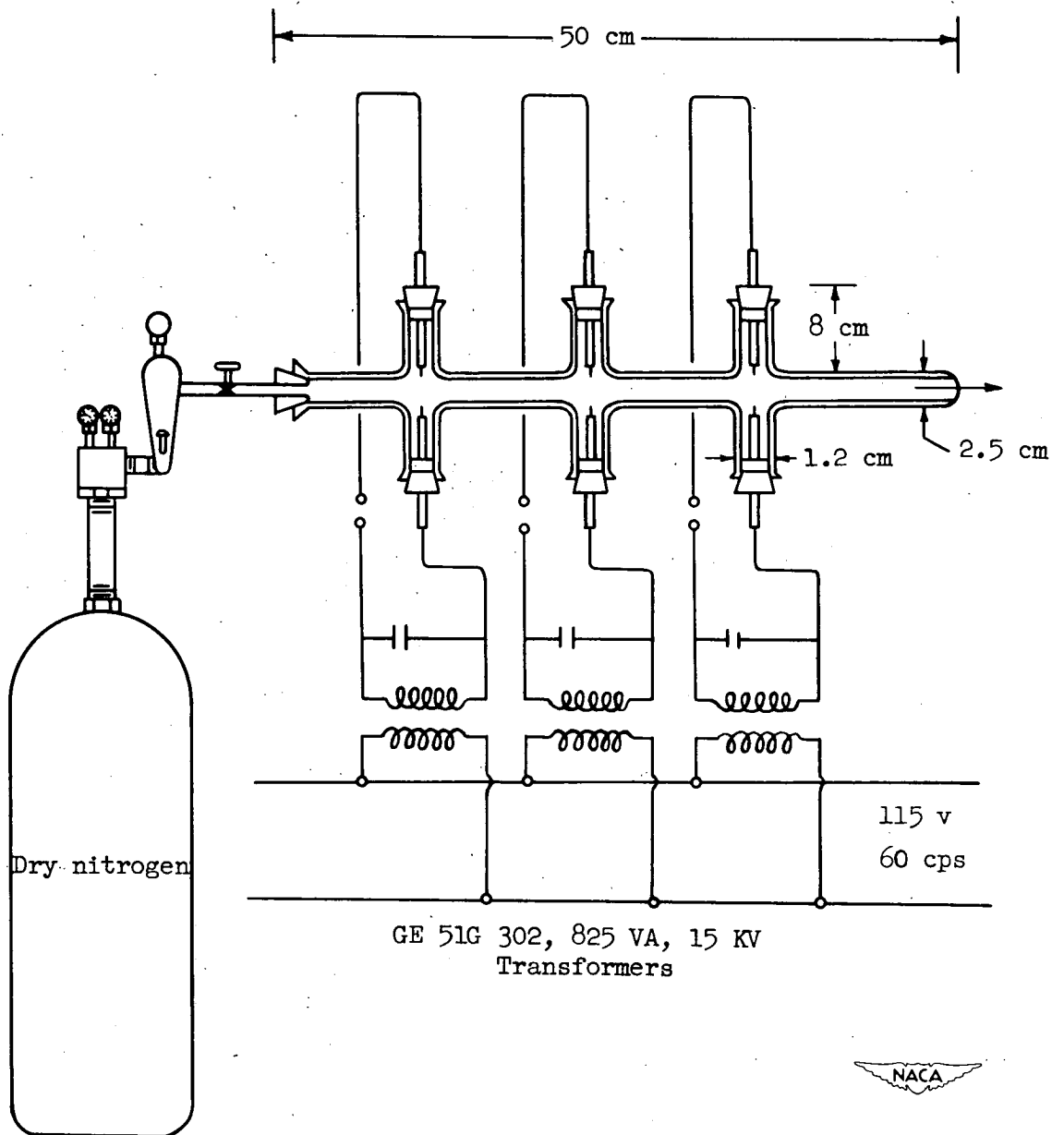


Figure 13.- Diagram of apparatus for exciting the stream of active nitrogen.



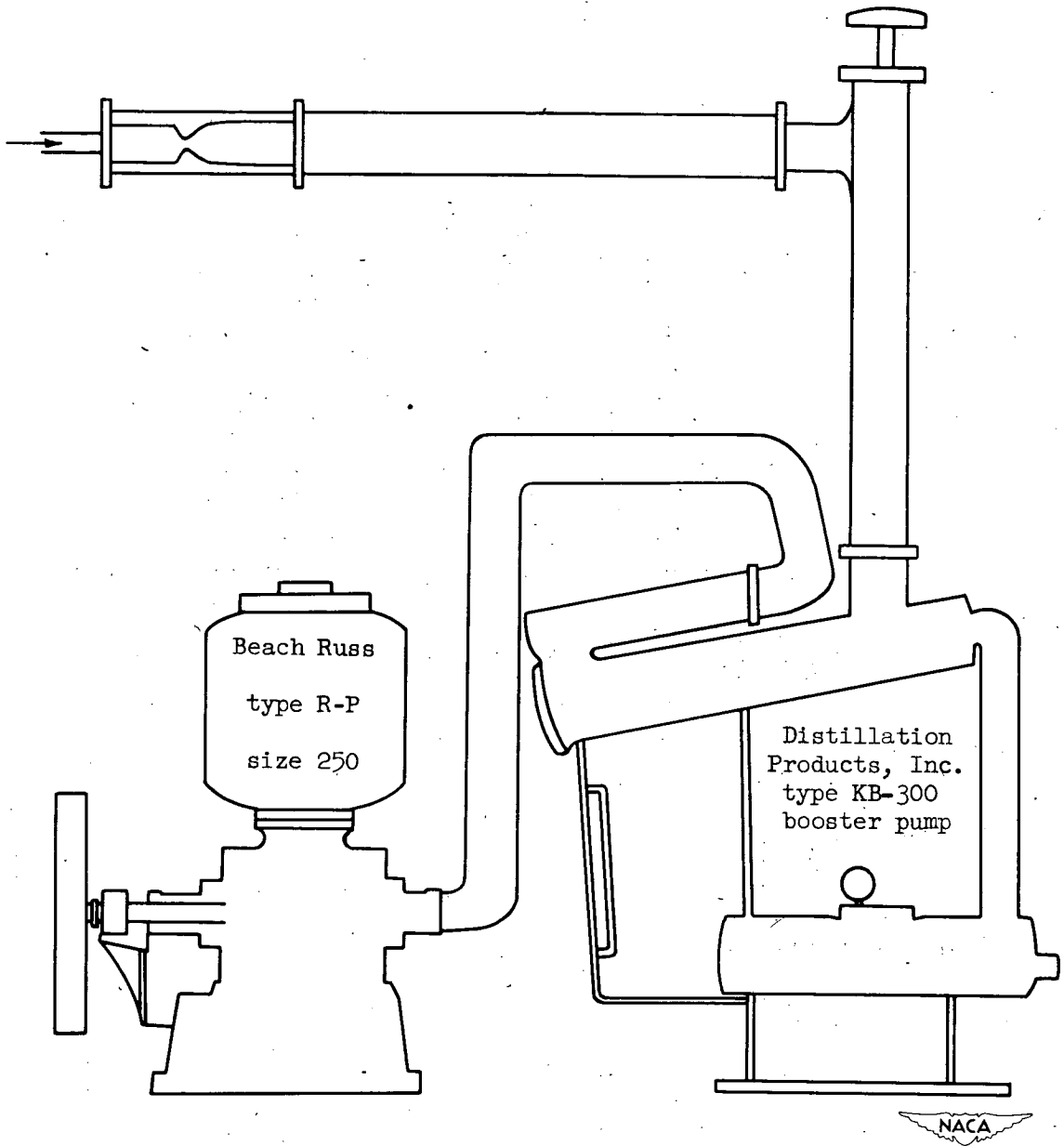


Figure 14.- Sketch of nozzle and pumps.

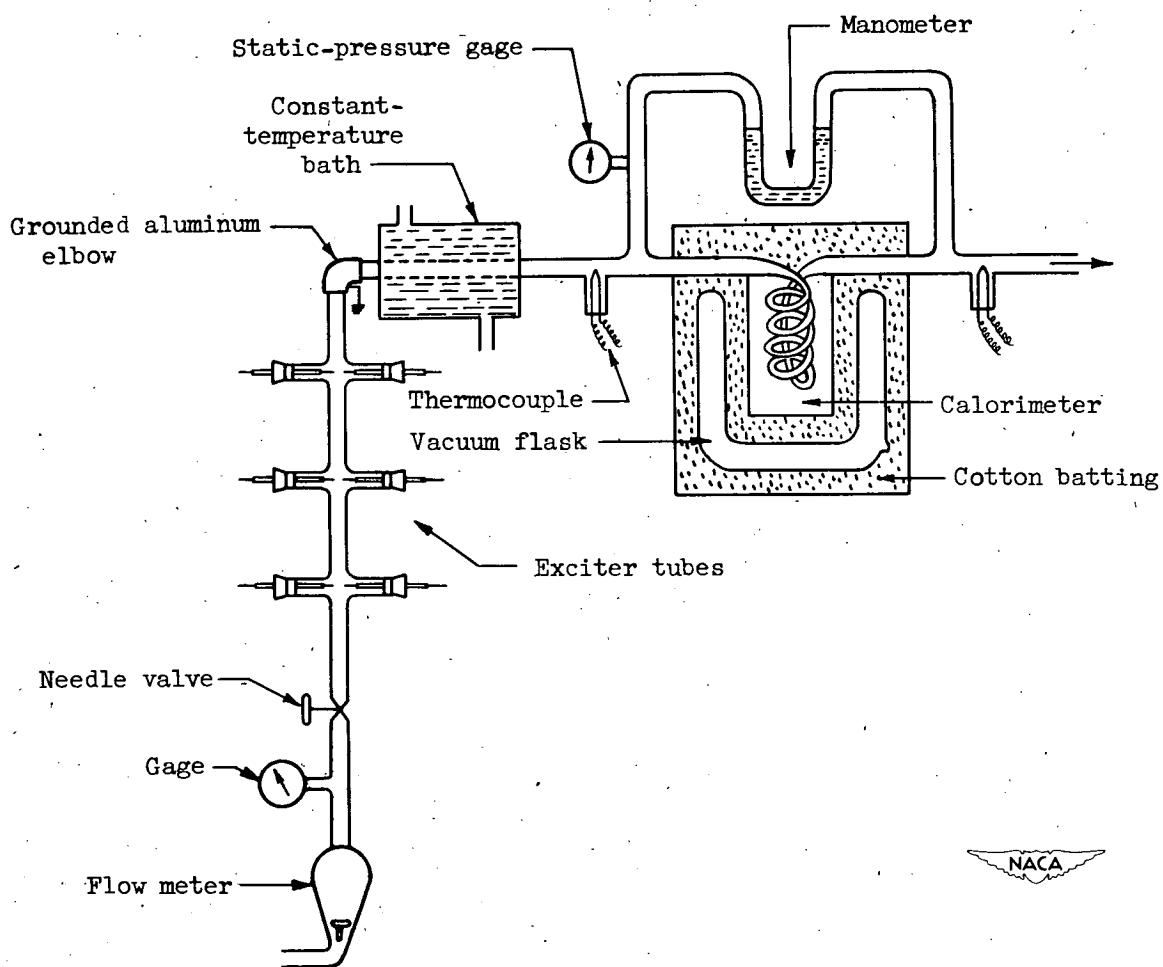


Figure 15.- Arrangement of viscosimeter and calorimeter.

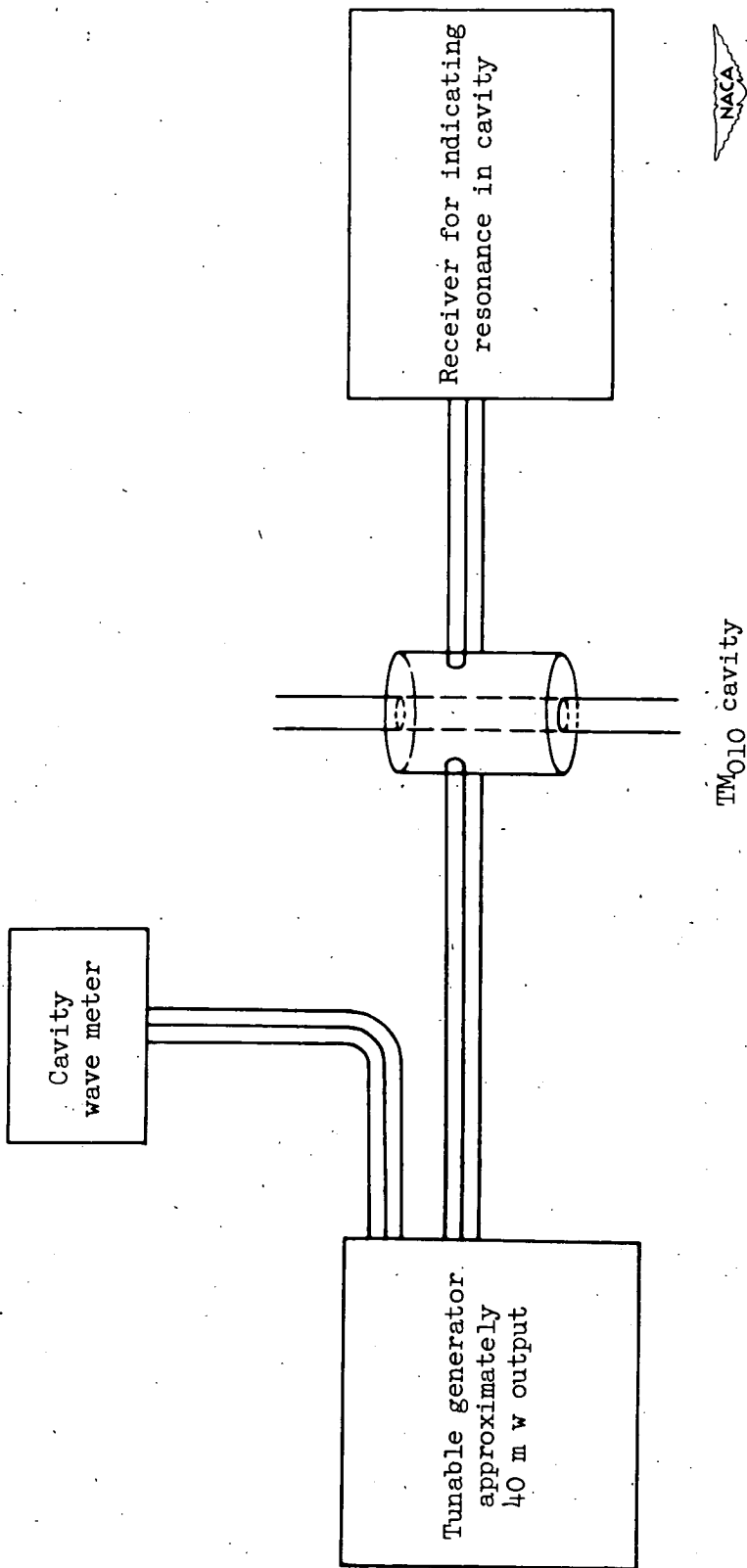


Figure 16.- Arrangement of apparatus for microwave measurements of electron density.

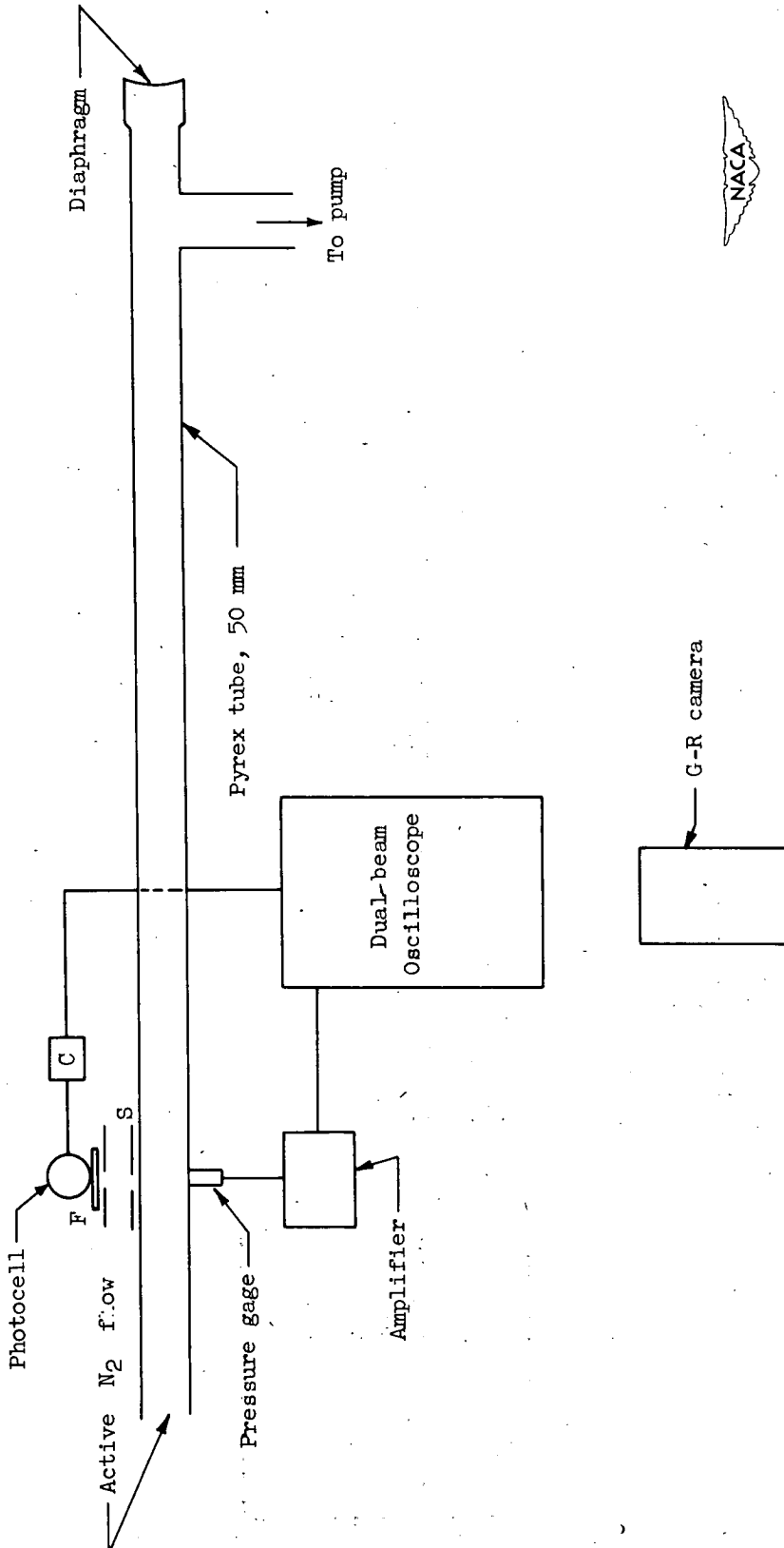


Figure 17.- Arrangement of shock tube and associated apparatus.  
F, filter; S, collimating slits; C, cathode follower.

Flow of Rarefied Gases

1.1.5

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The Physical Properties of Active Nitrogen in Low-Density Flow.

By James M. Benson

NACA TN 2293

February 1951

(See Abstract on Reverse Side)

Research Technique, Aerodynamics

9.2.2

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Benson, James M.

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(See Abstract on Reverse Side)

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### Abstract

An investigation of the physical properties of active nitrogen in low-density flow indicates that active nitrogen may be used for flow visualization without introducing excessive disturbances in the stream. Quantitative measurements of density in the flow may be obtained from certain restricted regions of the spectrum of the afterglow. The results are shown to disagree with Mitra's theory of the afterglow. The results appear to require that active nitrogen contain metastable molecules. Methods are described for the use of active nitrogen in shock tubes.

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