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## Abstract

This report summarizes project activity during the period 1 April 1970 to 30 June, 1970. Modifications of the instrumentation used for high resolution measurements of  $15\mu\text{m}$   $\text{CO}_2$  and the analysis of initial spectra which led to these modifications are described. Details of the construction and testing of components of the ozone generating, handling and measuring system are discussed. Initial calculations and development of a computer code for the line positions of  $9.6\mu\text{m}$  Ozone are received. Reports published and papers presented are noted.

## I. Introduction

This is the 3rd Quarterly Progress Report on Contract No. NSR 23-005-376, covering the period 1 April 1970 to 30 June 1970.

The project effort during this time period was divided among the following tasks.

- A. High resolution measurements of the  $15\mu\text{m}$  absorption band of  $\text{CO}_2$  (L. W. Chaney).
- B. Theoretical analysis of high resolution  $15\mu\text{m}$   $\text{CO}_2$  measurements (S. R. Drayson).
- C. Development of ozone generating, handling and measuring system (L. T. Loh and P. A. Titus).
- D. Theoretical analysis of  $9.6\mu\text{m}$  ozone band (W. R. Kuhn).
- E. Report Writing

## II. High Resolution Measurements of the $15\mu\text{m}$ Absorption Band of $\text{CO}_2$

(L. W. Chaney)

At the beginning of this reporting period the Jarrell-Ash 1.8 meter spectrometer was completely set up and ready for the study of  $15\mu\text{m}$   $\text{CO}_2$  absorption.

Spectra were then taken with the 8.74 cm cell at various pressures between 5 mm and 640 mm in the wavelength regions  $672\text{ cm}^{-1}$  to  $677\text{ cm}^{-1}$ ,  $696\text{ cm}^{-1}$  to  $701\text{ cm}^{-1}$ ,  $711\text{ cm}^{-1}$  to  $717\text{ cm}^{-1}$  and near the Q branch of the isotope  $\text{C}_{12}\text{O}_{16}\text{O}_{18}$  at  $662\text{ cm}^{-1}$ . The Analysis of the data has been carried out by S. R. Drayson (see section III). It was decided that further modifications were necessary to improve the instrument characteristics.



A new baffle was designed and installed in the spectrometer (see figure 1). The new baffle extends across the chamber and is located between the slits and the pass mirrors. It consists of two rectangular holes in a flat plate. The direct beam from the main mirror and the 'first pass' light which passes over the top of the exit pass mirror are prevented from reaching the exit slit. There is a portion of the beam near the top of the exit pass mirror that contains both the first pass and second pass light. The baffle removes all of this light and thus reduces the total signal.

The mirror mount for the horizontal ellipse in the exit optics was modified to reduce its sensitivity to vibration. New mounting posts were made to locate the mirror more precisely and the basic mount was reduced in size to insure clearance from the outside shell.

The main vacuum pump was bolted to the concrete floor to reduce the instrument vibration. Also, its location was changed in order to provide space for the location of the 40 meter cell which will be used at a later date.

A high vacuum valve was installed between the chamber and the pump to allow for filling the main chamber with gas for long path studies.

After the above noted modifications were completed, the instrument was reassembled and aligned. A test spectrum indicated that:

1. The stray light was essentially reduced to zero.
2. The instrument function is very close to the ideal  $\sin x/x$  shape.
3. The resolution appears to be slightly improved.
4. The signal to noise ratio is worse by a factor of two.

The decrease in signal was expected as a result of baffling the first pass light. The baffling also reduced the final slit height. A careful

measurement indicated that the final image was slightly less than 2 mm., whereas the detector is 4 mm. high. Hence it has been decided to mass the flake at the detector to reduce the noise. A reduction in noise will also be optimizing the preamplifier input resistance. It is expected that the noise can be reduced to 70% of its present value by these techniques.

It is planned that spectra previously recorded will be repeated. Also spectra will be taken with other cells 0.5 cm, 2.5 cm and 5.0 cm long. It is also expected that some nitrogen broadened spectra will be obtained. It is expected that near the end of September that the transfer optics required for the installation of a 40 meter cell will be installed.

### III. Theoretical Analysis of High Resolution $15\mu\text{m}$ $\text{CO}_2$ Measurements

(S. R. Drayson)

The computer programs to handle the card output of spectra from the Jarrel-Ash instrument were revised to give improved normalized plots of the experimental data.

1) The programs were modified to plot simultaneously the theoretically computed spectra for conditions corresponding to those of the experimental data. This procedure is very useful for quick checks on the laboratory spectra to compare wavenumber calibration and to estimate resolution. It also provides a rough estimate of the accuracy of the parameters used in the calculation. Two examples of such plots are shown in figures 2 and 3.

Figure 2 shows a region containing the Q-branch of the band (010:1-100:0). It can be seen that the theoretically computed wavenumbers of the high J-number lines of the Q-branch (Q54-58) are slightly too low, indicating a need for revision of the constants from which they are calculated. A similar situation exists for a number of bands shown in Figure 3.

2) Although the noise level shown in Figure 1 is acceptable, an improvement can easily be made. This is important if the slits are further closed to attempt to obtain a higher resolution.

Part of the noise comes from noise in the background spectrum. Since its structure exhibits features that change only slowly with wave-number, it may be considerably smoothed without changing the accuracy or resolution of the line spectra. A five point triangular smoothing function was used.

In this spectral region the digitizer produces data points approximately  $0.01 \text{ cm}^{-1}$  apart, while the resolution of the spectrometer is approximately  $0.08 \text{ cm}^{-1}$ . Thus the line spectra were smoothed using a similar technique to that used for the background, but involving only 3 points. These two procedures considerably dampened the noise with only a slight loss of spectral resolution. The spectra in Figure 3 are examples of smoothed spectra.

A close examination of the spectra in Figure 3 shows an undesirable feature. The lines of the fundamental (IR 8 and IR 10) show saturation near the line centers for the three highest pressures in the theoretical calculation. The experimental spectra also show this feature but the level at which saturation occurs changes with increasing pressure. This was deduced to result from stray energy that was falling on the detector, due to inadequate baffling in the instrument. The modifications made to the spectrometer are described in another section.

In May 1970 some aspects of this work were described in a paper given at the Symposium on Remote Sounding of the Atmosphere at the COSPAR meeting in Leningrad, USSR. The title of the paper was 'Transmittances for use in remote soundings of the atmosphere' by S. R. Drayson. The paper

was sponsored jointly by the National Environmental Satellite Laboratory, ESSA and NASA, Goddard Space Flight Center.

IV. Development of Ozone Generating, Handling and Measuring System  
(L. T. Loh and P. A. Titus).

The construction and testing of component parts of the ozone generating, handling and measuring system has continued.

The Barocel pressure gage was received from the manufacturer. It was put through a series of calibrations relative to a Cenco mercury manometer. To our surprise, the discrepancy found was much larger than could be accounted for by the limited resolution of the NLS digital voltmeter used to read the Barocel output. The factory, was contacted and the Barocel was shipped back for a calibration check. After a long wait and several telephone calls, the Barocel finally was returned. Mr. Sydney Locke, sales engineer, said that the original calibration at one point (.777) was in error. A new value (.805) was given. The calibration was checked again and found to be satisfactory.

The Heise stainless steel Bourdon tube manometer was received. It was compared with the Cenco manometer. The material is type 316, known for good chemical resistance but for poor spring property. The zero point shift is 2 to 4 times that of good Bourdon material, such as beryllium copper. With this limitation in mind, the gage performed quite well.

The idea of using conventional stove pipe to exhaust the ozone to the outside of the laboratory was conceived. After some discussion the installation was made and was successful.

The Jarrel Ash monochromator system also arrived during this quarter without the scan drive motor, which came in 37 days later. The instruction manuals provided were inadequate, particularly those for the photomultiplier power supply and amplifier. The descriptions of the amplifier's analog signal in these two manuals contradicted one another. Otherwise, the uv light source, the monochromator, the detector and its electronics were satisfactory. The wavelength calibration was within specification and the output signal ample. A 1 P 28 phototube (far less expensive) was substituted for the factory detector tube and got more than enough output.

The uv light will be used to monitor ozone in the White cell. Transfer optics are required to direct the uv light beam through the White cell and across 2 vacuum barriers into the monochromator and detector. A proposed design was checked on the optical bench and transformed into hardware. The transfer optics was mounted on the white cell. The arrangement of the apparatus is shown in figure 4. The two lenses are made of quartz and formed vacuum tight seals. The entire uv system was checked. All 6 mercury lines were obtained in the spectrum and each produced enough output.

The scan drive motor arrived late. It was mounted on the monochromator and tested. The first time it was used, it caused trouble. A stall clutch allegedly mounted between the motor and the lead screw failed to work. The motor overdrove the lead screw of the monochromator causing loss of wavelength calibration. The factory was consulted. The service department suggested the return of the instrument for servicing.

A good hard look at the scan mechanism showed that the cause of the failure was due to the loosening of one set screw. It was put back and the wavelength calibration was set back in place. The monochromator was checked once again for calibration.

A gas stirrer was needed for the White cell. Difficulties encountered in the design were the chemical activity of ozone and the existence of a vacuum barrier. It was decided to use ozone-proof materials and a magnetic drive. The drive worked easily across the White cell wall. Upon testing it was found that the magnetic drive was strong enough to turn the mixer blade across the wall, but the bearings failed due to too much sidewise play. Modifications of the design are being considered.

The air filters and air dryer originally on the wall of the spectrometer room were found to be not in working order. The air filters were cleaned and the air dryer serviced. The dry air has been used to dry a batch of spent silica gel, and also to keep the Perkin-Elmer 221 dry. The system was turned off during the weekend. When next used, the compressed air line was allowed to vent into the room to get rid of the water accumulated in the line. The system was then put into operation. This time, a large quantity of water appeared in the air filters (365 grams in 5 hours). The line was immediately stopped and the water removed. It is dangerous to operate the air dryer with this kind of moisture-laden compressed air. Modifications of this system are also being considered.

The Perkin-Elmer 221 was turned on for two working days and a few polystyrene spectra were recorded.

V. Theoretical Analysis of 9.6 $\mu$ m Ozone Band (W. R. Kuhn)

Work on the ozone molecule has continued during the last report period. The major effort has been directed toward developing a computer code for the line positions in the 9.6 $\mu$ m region. Numerical calculations to verify the theoretical development of the problem have been carried out; for example, for the simple case of rotational level J=1 for which there will be three levels  $1_{01}$ ,  $1_{11}$ ,  $1_{10}$ , the energy matrix E(K) for the  $I^r$  representation is,

$$\begin{array}{c|ccc}
 K & -1 & 0 & 1 \\
 \hline
 -1 & 1/2(K+1) & 0 & -1/2(K+1) \\
 0 & 0 & K-1 & 0 \\
 1 & -1/2(K+1) & 0 & 1/2(K+1)
 \end{array}$$

where K is the asymmetry parameter. This matrix can be diagonalized by the Wang transformation which has the form.

$$X = \frac{1}{2} \begin{vmatrix} -1 & 0 & 1 \\ 0 & 2 & 0 \\ 1 & 0 & 1 \end{vmatrix}$$

and

$$E'(K) = X^T E X = \begin{vmatrix} K+1 & 0 & 0 \\ 0 & K-1 & 0 \\ 0 & 0 & 0 \end{vmatrix}$$

which represents the three energy values for  $1_{10}$ ,  $1_{01}$ , and  $1_{11}$ . One can also express the energy in terms of the submatrices  $E^+$ ,  $E^-$ ,  $0^+$  and  $0^-$ , one for each of the symmetry species in the V group. For our particular example these are easily shown to be  $E^+ \rightarrow K-1$ ,  $0^- \rightarrow K+1$ , and  $0^+ \rightarrow 0$ .

From the symmetry calculation for J odd (1),  $E^+$  is associated with  $1_{01}$  of energy K-1,  $0^-$  with  $1_{10}$  of energy K+1 and  $0^+$  with  $1_{11}$  of energy zero.

A similar example was carried out with  $J=2$  and the method of continued fractions was used. Work is continuing in the development of a computer code for this method.

Work has also begun on a comparison of middle-atmosphere flux divergences in the  $9.6\mu\text{m}$  region between Clough and Kneizy's (1966) band parameters and those of Kaplan, Migeotte and Neven (1956). Recent preliminary results between theoretical calculations of London (unpublished) and experimental work of Hayes and Roble (unpublished) indicate good agreement in middle-atmosphere ozone concentrations. Better ozone data justify a more critical study of the transmission function and this comparison will reflect the importance of uncertainties in the band parameters on the  $9.6\mu\text{m}$  flux divergence calculations.

During this report period, a review paper on the Radiative Processes and Radiative Sources and Sinks in the Middle Atmosphere was also presented to the AMS symposium on the Dynamics of the Mesosphere and Lower Thermosphere in Boulder Colorado on June 15-18. This paper will appear in a separate report.



## VI Reports Published

A paper "Transmittances for use in Remote Soundings of the Atmosphere," by S. R. Drayson was presented at the Symposium on Remote Sounding of the Atmosphere at the COSPAR meeting in Leningrad, USSR in May.

Papers were presented by 2 members of the laboratory at the Symposium on the Dynamics of the Mesosphere and Lower Thermosphere of the AMS, June 15-18, 1970, Boulder, Colorado. The papers were:

1. Review paper: Radiation processes and the distribution of radiative sources and sinks. William R. Kuhn.
2. Carbon dioxide long-wave radiative transfer for use in models of the middle atmosphere. S. R. Drayson.

The PhD dissertation "The Measurement of Atmospheric Ozone Using Satellite Infrared Observations in the 9.6 $\mu$ m Band," by James Madison Russell III was published as report No. 03635-1-T. The work was supported in part by this contract and in part by funds on NASA contract NGR 23-005-394 and by funds from ESSA under contract E 92-69-N.

## VII References

1. Clough, S. A. and F. Y. Kniezys, 1965: Ozone Absorption in the 9 Micron Region, Report, AFCRL 65-862, November 1965.
2. Kaplan, L. D., M. V. Migeotte, and L. Neven, 1956: 9.6 Micron Band of Telluric Ozone and its Rotational Analysis, J. of Chem. Phys. 24, pp. 1183-1186.

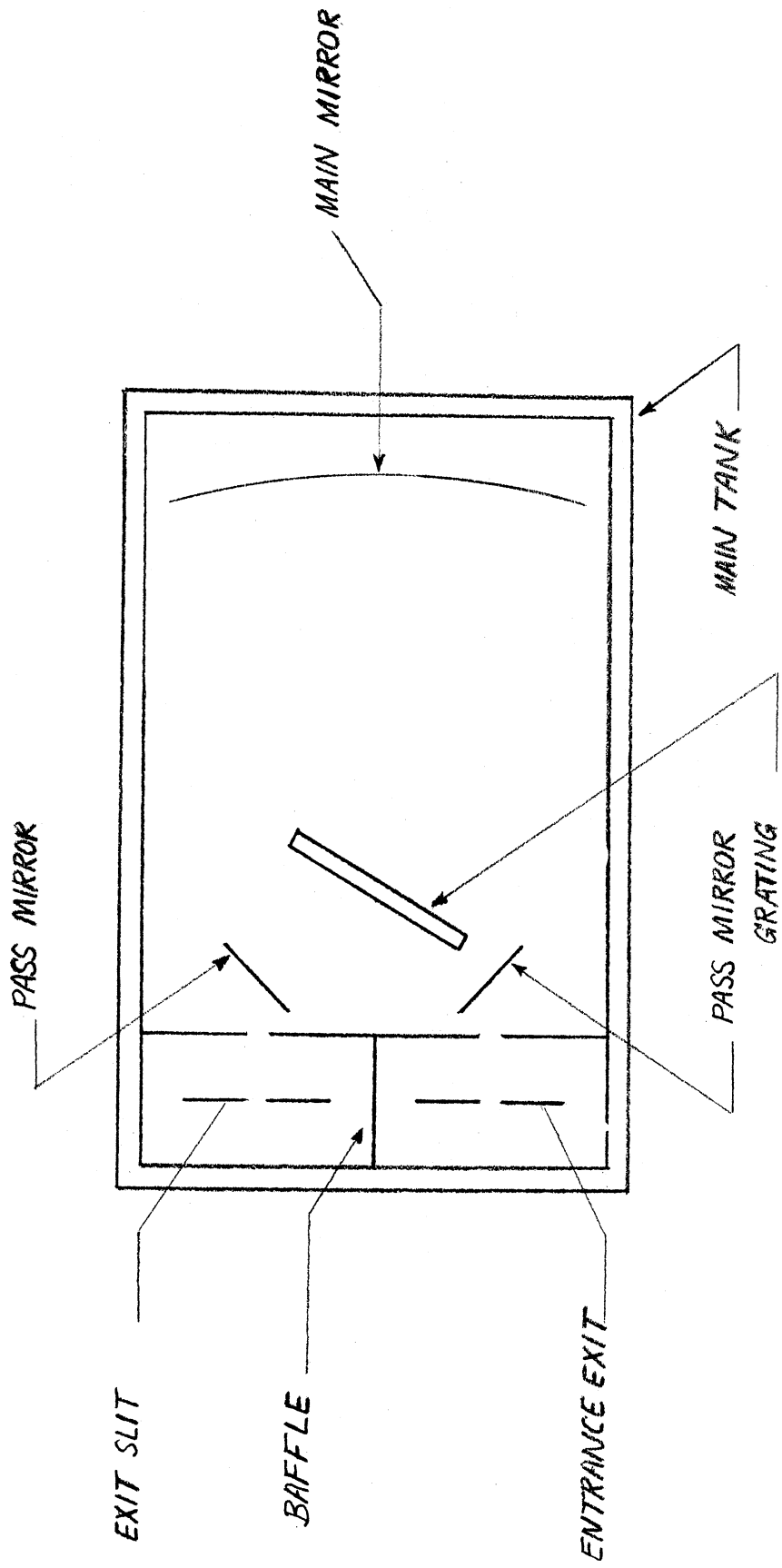


Figure 1. Baffle for 1.8 meter Jarrel Ash spectrometer

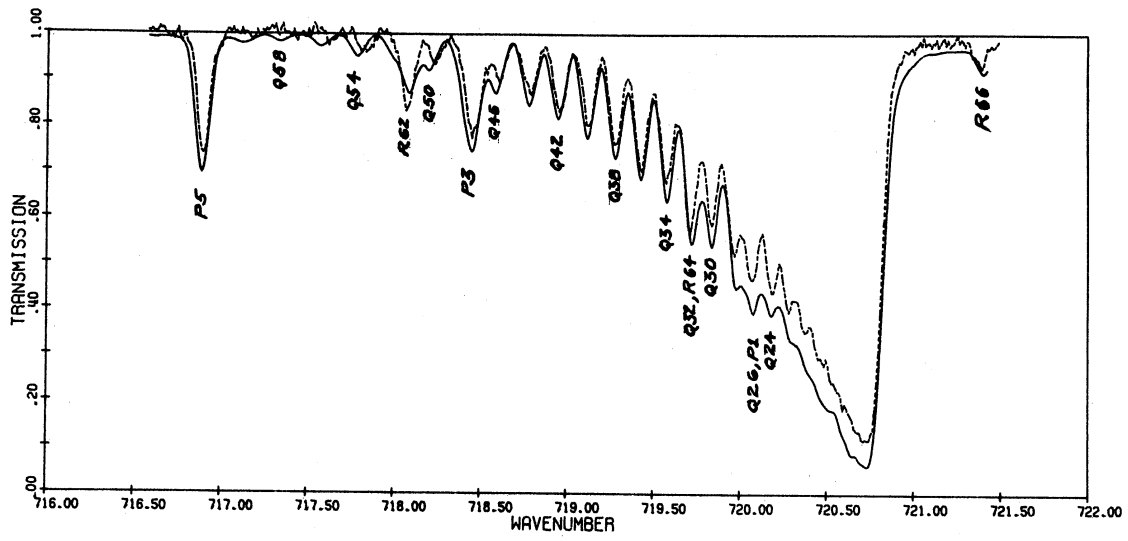


Figure 2. Comparison of high resolution spectrum (dashed lines) with theoretically computed spectrum (solid lines) of pure carbon dioxide. Theoretical resolution is  $0.08 \text{ cm}^{-1}$ . All identified lines belong to the P- or Q- branches of (010:1-100:0) band except lines of R-branch of the  $\nu_2$  fundamental (000:0-010:1) band. Cell length=8.74, pressure 75.08 torr, temperature  $25^\circ \text{C}$ .

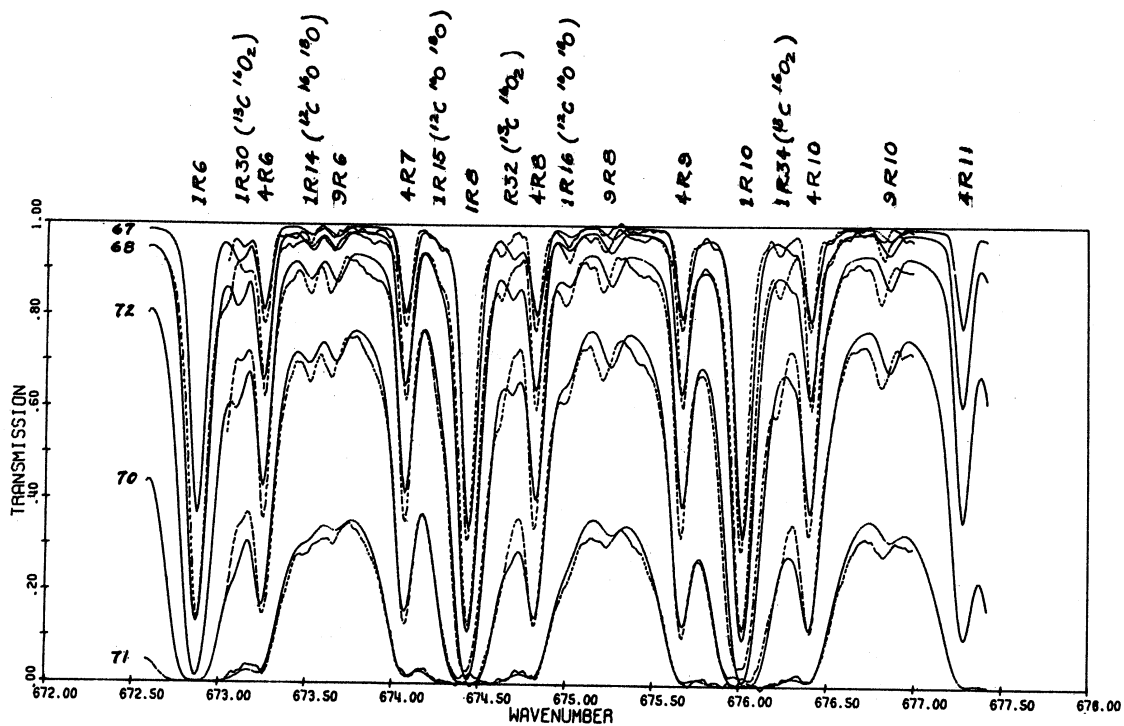


Figure 3. Comparison of high resolution spectra (dashed lines) with theoretically computed spectra (solid lines) of pure carbon dioxide. Theoretical resolution is  $0.08 \text{ cm}^{-1}$ . Band code is:  
 Band 1 (000:1 - 010:1)  
 Band 4 (010:1 - 020:2)  
 Band 9 (020:2 - 030:3)  
 Cell length = 8.74 cm, temperature  $26^{\circ}\text{C}$ . Pressures 20.01, 40.04, 80.19, 159.98 and 320.01 torr.

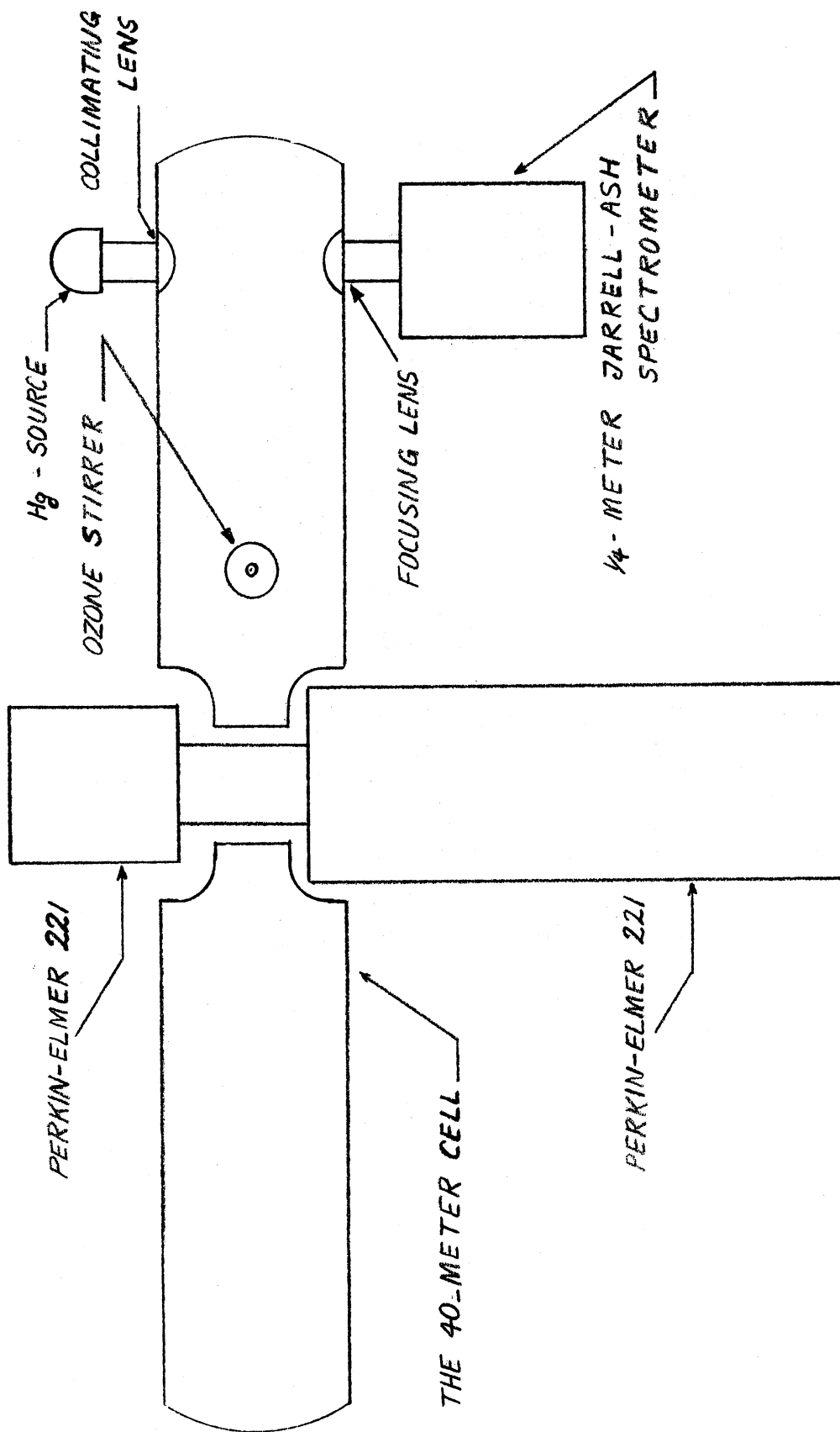


Figure 4. Schematic diagram showing method of measuring ozone with Jarrell Ash 1/4 meter spectrometer.

