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ON THE MEASUREMENT OF RAYLEIGH SCATTERING BY GASES AT 6328Å

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16. Abstract

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SUMMARY

The problem of laboratory measurements of Rayleigh scattering and depolarization ratio for atoms and molecules in the gaseous state is described. It is shown that, if the scattered radiation measurements are made at two angles, 90° and 54°44', the normal depolarization ratio cannot be determined meaningfully. However, from scattering measurements at 54°44', the Rayleigh scattering cross sections can be determined accurately. The measurements of Rayleigh scattering from He, H_2, Ar, O_2, and N_2 for unpolarized radiation at 6328Å are reported for 90° and 54°44' and compared with similar measurements at 6943 and 1215.7Å.

INTRODUCTION

The measurements of Rayleigh scattering and depolarization ratios for gaseous atoms and molecules have been reported by many investigators (refs. 1-13). These measurements were made at selected wavelengths in the visible region. Besides, there are only a few

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measurements in the vacuum ultraviolet (refs. 14-17). Experimentally to the best of our knowledge, this problem of Rayleigh scattering and the depolarization ratio has not been investigated as a function of wavelength. Conventionally, the Rayleigh scattering cross sections are calculated from the index of refraction at a wavelength and then extrapolated to other wavelengths using inverse λ^4 law (refs. 18, 19). The earlier measurements were limited in scope because of the experimental difficulties concerning the intensity of the light source, bandwidth, collimation, and above all the lack of instrumentation for detecting the feeble scattered signals. In recent years, the availability of intense light sources, especially lasers and the more sophisticated electronic equipments, have provided a powerful tool not only to study the phenomenon of Rayleigh scattering but still weaker interaction known as Raman scattering. Several investigators (refs. 20-27) have utilized these means for measuring the photon scattering (Rayleigh and Raman) and depolarization ratios for atoms and molecules. There are discrepancies in the measurements, particularly in the values of the depolarization ratios. In view of the importance of the Rayleigh and Raman scattering to the problems of atmospheric optics in general and to the remote sensing and other applications for planetary missions in particular, it was decided to look into the very technique of measuring Rayleigh scattering and depolarization ratio as a function of angle. Some of the important conclusions are presented and the results for 90° and 54°44' scattering from He, H_2, Ar, O_2, and N_2 are reported and compared with similar measurements at other wavelengths.

PROCEDURE

According to classical electromagnetic theory, the photon flux, Φ_θ, scattered by the gas molecules of number density N per unit solid angle at an angle θ from an unpolarized incident photon beam of intensity Φ_o is given by (refs. 14, 16, 28)

$$\Phi_\theta = \Phi_o \frac{3N}{16\pi} V_\theta \sigma_s \frac{2}{2 + \rho_n} \left(1 + \cos^2 \theta + \rho_n \sin^2 \theta\right) \qquad (1)$$

where V_θ is scattering volume; σ_s and ρ_n are the Rayleigh scattering cross section and normal depolarization ratio of the molecule, respectively. Heddle (ref. 14) and Samson (ref. 28) have suggested that if the scattered radiation measurements are made at two angles, namely, 90° and 54°44', the values of scattering cross section and the normal depolarization ratio for gas molecules can be determined more accurately. Let us examine this technique with respect to scattering cross sections and depolarization ratios as follows:

Scattering Cross Sections

The method commonly employed for measuring the Rayleigh scattering is to observe the scattered signal at right-angle to the incident radiation. However, if the scattered radiation, Φ_{54}, is observed at an angle 54°44' from the scattering volume V_{54}, then it can be shown from the equation (1) that

$$\Phi_{54} = \Phi_o \frac{V_{54}}{4\pi} \sigma_s N . \qquad (2)$$

From equation (2) it is obvious that the measurements of Φ_{54} as a function of number density will provide a straight line, the slope of which will give the value of the scattering cross section. It may be remarked that the equation (2) is independent of ρ_n Therefore, the scattering cross section is not affected by the uncertainty in the ρ_n value. The values of ρ_n are wavelength dependent and to the best of our knowledge have not been measured as a function of wavelength for any of the gases. The ρ_n values have been measured by many investigators (refs. 3, 6, 7, 9-13, 29) at selected wavelengths in the visible region and are generally assumed to be constant for all wavelengths. However, this assumption may not be valid in the vacuum ultraviolet region where the measurements are not known. Even in the visible region there are discrepancies between values obtained by various investigators (refs. 3, 6, 7, 9-13, 29). Some of the gases used in these measurements may not be free from dust particles which might have introduced some error in the measurements. Also, most of the measurements were made for total scattered intensity which is the sum of the Rayleigh and Raman components. It is believed that the Raman components are strong polarizers and consequently might have introduced some error. The recent work of Rowell et al (ref. 29) has shown that by eliminating the Raman components, the values of ρ_n are found to decrease. In the light of the foregoing discussion, it seems that the measurements of scattered light at 54°44' being independent of ρ_n value may yield more accurate results of scattering cross sections compared to those which are obtained from the measurements at any other angle. The numerical accuracy of the results obtained from such measurements is discussed in the Experiment and Results Section. (Also, see Appendix A.)

Depolarization Ratios

Let us assume that Φ_{90} is the scattered intensity at an angle 90° from the scattering volume V_{90}; then from equation (1),

$$\Phi_{90} = \Phi_o \frac{3N}{16\pi} V_{90} \sigma_s \frac{2(1 + \rho_n)}{2 + \rho_n} \tag{3}$$

By the combination of equation (2) and (3), we get

$$\frac{\Phi_{54}}{\Phi_{90}} = \frac{2}{3} \frac{V_{54}}{V_{90}} \left(\frac{2 + \rho_n}{1 + \rho_n}\right) , \tag{4}$$

or

$$\rho_n = \left[4 - 3 \left(\frac{V_{90}}{V_{54}} \frac{\Phi_{54}}{\Phi_{90}}\right)\right] \Bigg/ \left[\left(3 \frac{V_{90}}{V_{54}} \frac{\Phi_{54}}{\Phi_{90}} - 2\right)\right] . \tag{5}$$

For the purpose of determining the ratio V_{90}/V_{54}, let us consider the geometry of the experimental setup used in the measurements which are presented in the Results Section. In this setup the incident radiation is a parallel beam of light whereas the scattered radiation forms a converging beam at the detector. This type of setup is most commonly used and its sectional diagram is shown in Figure 1. It is clear from the figure that scattered radiation is observed at the angles of 90° and θ° with respect to incident radiation. If A_{90} and A_θ are the areas of intersection of the bisecting planes of incident and scattered radiations corresponding to angles 90° and θ°, then it may be assumed that $V_{90}/V_\theta \approx A_{90}/A_\theta$. From the figure,

$$A_{90} = \text{Area } \overline{ABCD} = YZ \cdot MP = 2YZ \cdot NO \cdot \tan \alpha \tag{6}$$

$$\text{Similarly, } A_\theta = \text{Area } \overline{EFGH} = YZ \cdot LQ = 4YZ \cdot NO \cdot \sin \alpha \frac{\sin \theta \cos \alpha}{\cos 2\alpha - \cos 2\theta} . \tag{7}$$

$$\text{Therefore, } \frac{V_{90}}{V_\theta} \approx \frac{A_{90}}{A_\theta} = \frac{\cos 2\alpha - \cos 2\theta}{2 \sin \theta \cos^2 \alpha} . \tag{8}$$

From expression (8) it can be shown that V_{90}/V_θ is not unity. For an ideal experimental setup in which both the incident and scattered radiations form the parallel beams of light, the angle α is equal to zero. Hence, for such a case

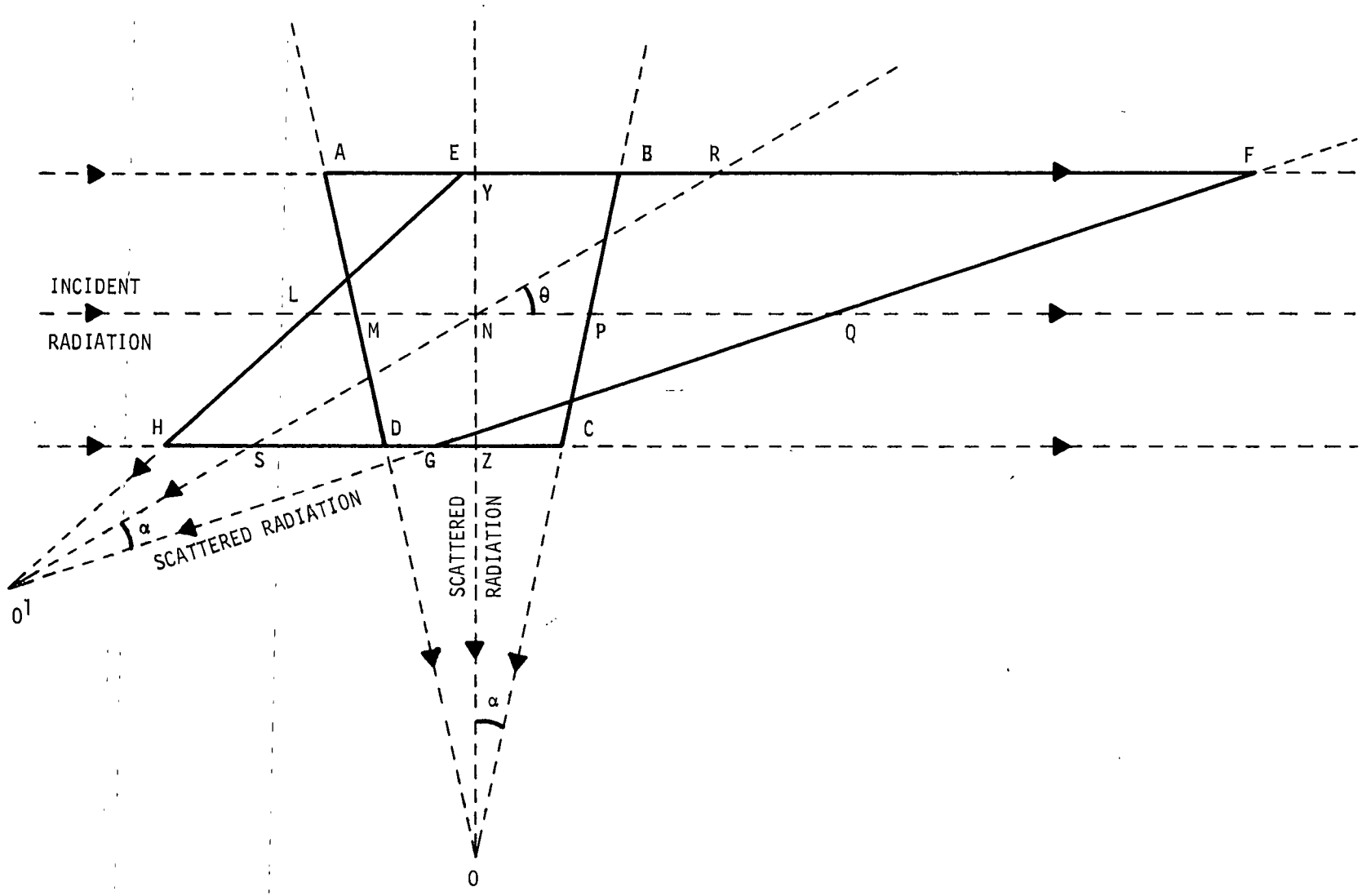

Figure 1. The sectional diagram of the incident and scattering radiation.

$$\frac{V_{90}}{V_\theta} = \sin\theta \, . \qquad (9)$$

It is obvious from expression (9) that V_θ is always greater than V_{90} except for $\theta = 90°$ when V_θ becomes equal to V_{90}. For $\theta = 54°44'$, $V_\theta = 1.225\ V_{90}$. Therefore, it may be realized that in most commonly used experimental arrangements the scattering volumes at two different angles are not the same.

Now let us further examine whether the technique, proposed by Heddle (ref. 14) and later re-emphasized by Samson (ref. 28), can be used meaningfully for the determination of ρ_n values. As seen from equations (4) and (5), one needs to measure the scattered radiation at two angles, namely 54°44' and 90° for obtaining the value of ρ_n. It can be shown that the value of ρ_n determined from equations (4) and (5) is very sensitive to the measurement of Φ_{54}/Φ_{90}. The following example will illustrate this point. The values of the depolarization ratio are very small for most of the gases (ref. 29; $\rho_n{}^{N_2} = 0.0214$; $\rho_n{}^{O_2} = 0.0565$; $\rho_n{}^{H_2} = 0.0188$; $\rho_n{}^{CO} = 0.0117$). Let us take the value of $\rho_n = 0.0214$ for nitrogen and $V_{90}/V_{54} = 0.816$; then to satisfy the equation (4) the value of Φ_{54}/Φ_{90} will be 1.615. If we assume that 5 per cent is the typical error in the laboratory measurements of Φ_{54}/Φ_{90}, then the corresponding error in the value of ρ_n will be about 500 per cent. Even with an upper limit of $\rho_n = 0.5$ (ref. 30), the error in ρ_n value will be about 50 per cent corresponding to the same error of 5 per cent in the observations. Therefore, it may be concluded that this experimental technique cannot be employed meaningfully as it introduces about 10 to 100 times more error in a parameter to be determined compared to the error in observations (see Appendix B). In view of this discussion we would like to emphasize that the conventional method (ref. 29), in which the measurements made at one angle (for example, 90°) are utilized to determine the depolarization ratio, should be used. However, this conventional method cannot be used in the region of vacuum ultraviolet where the polarizers and the analysers are not available. Therefore, in absence of any other better technique, the technique of Heddle (ref. 14) and Samson (ref. 28) may be used keeping in mind that the ρ_n values thus determined would be in large error, 10 to 100 times the actual values.

EXPERIMENT AND RESULTS

The general configuration of the experiment is shown in Figure 2. It consists of a scattering chamber made of stainless steel which can withstand pressures up to 100 atmospheres. There are four ports in the chamber sealed with sapphire windows.

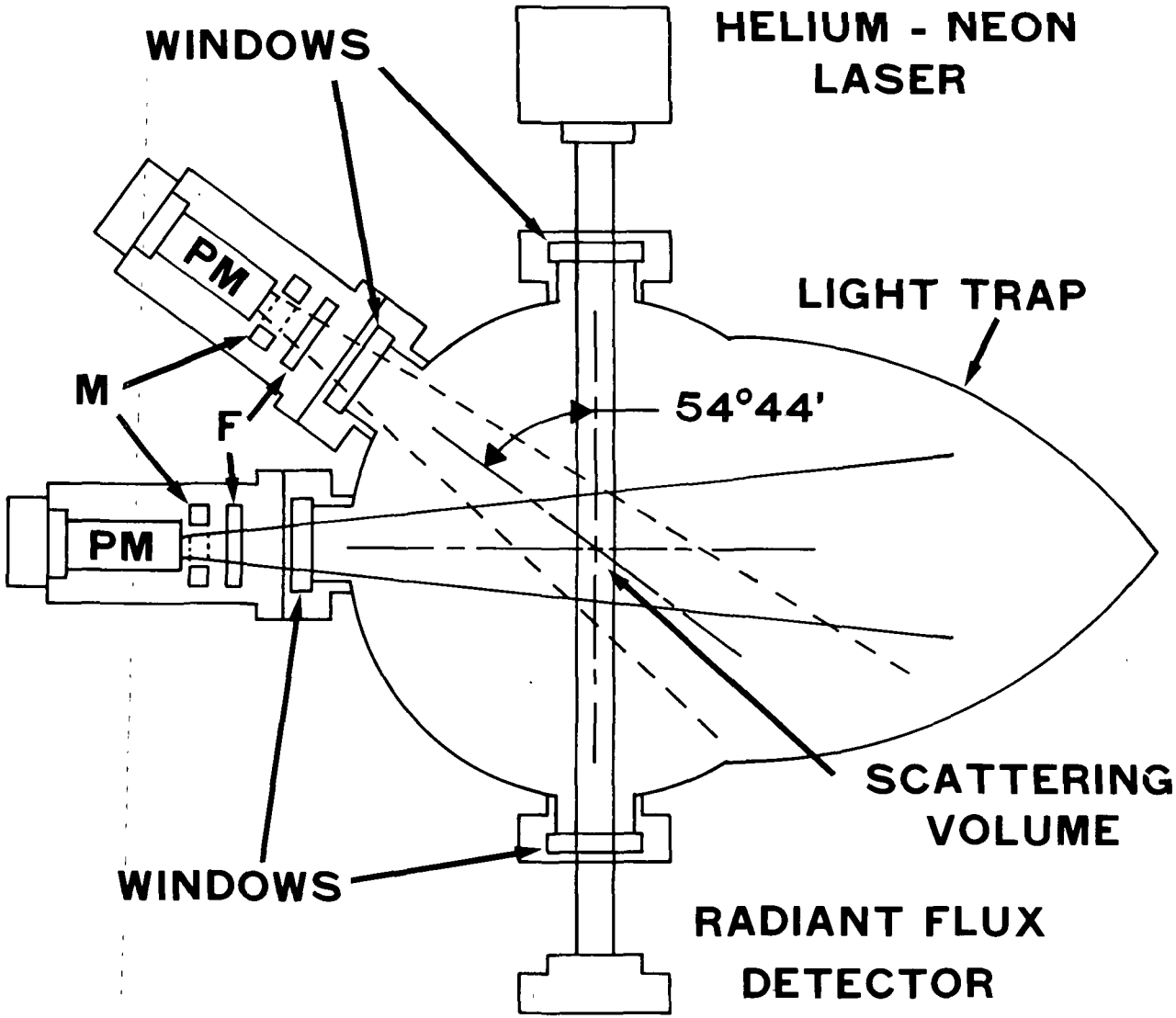

Figure 2. Apparatus for measuring scattered radiation at 90° and 54° 44'. F is a bandpass filter and M is a magnetic defocussing assembly used for the enhancement of signal-to-noise ratio.

The location of ports is as shown in the figure. There is also a light trap opposite two ports which hold the photomultiplier for measuring the light signal scattered at 90° and 54°44'. The inside surface of the chamber is coated with flat-black-chrome-finish that provides the dark background and thus reduces any spurious radiation reaching the detector. There are other ports, not shown in the figure, which are used for gas inlet, gas outlet, pumping, safety valve, and the pressure gauge.

The light source used in the present measurements is a helium-neon laser (Spectra Physics Model 132) which emits unpolarized radiation at 6328Å. The output of the laser is monitored constantly for changes in radiation flux by means of a Radiant Flux Detector (HP Model 8334A) and Radiant Flux Meter (HP Model 8330A) and is recorded on a strip chart recorder. The photon counting technique is used for the measurement of scattered radiation. For this purpose, a photomultiplier tube (EMI 9558Q) enclosed in a housing (SSR 1151), an amplifier-discriminator (SSR 1120), a photon counter (SSR 1105), and a digital recorder (HP Model 5055A) modified to match the SSR system, are used. The high gas pressure in the chamber and a defocussing magnetic assembly in front of the photomultiplier tube are used to enhance the signal-to-noise ratio which is an essential requirement for measuring the extremely weak light signals. The use of the magnetic assembly alone improved the signal-to-noise ratio by about a factor of 8 in the present setup. A narrow bandpass filter (half width 8Å) is also used for minimizing the effect of the Raman components.

Research grade gases supplied by AIRCO were used in the present measurements. There was no attenuation of radiation at 6328Å in any sample of the gases. However, it may be pointed out that the attenuation of radiation by absorbing species, if present, could be accounted for as the path lengths from the scattering volume to the detectors were equivalent in the present setup (ref. 16). The gas samples were allowed to pass through a series of millipore filters (VSWP 047 00) which filtered the dust particles of size larger than 25 mμ. The use of these filters minimize the scattering due to dust particles which have high scattering cross section as compared to the Rayleigh scattering. The filtered gas samples were then allowed to enter into the scattering chamber. The gas pressures were measured by a Heise pressure gauge which has a range of 0-100 atm. The scattered signals were measured as a function of the gas pressure at the angles of 90° and 54°44'. It would be appropriate to mention that George et al (ref. 22, 23) noted a forward enhanced asymmetry in the observations for argon and xenon, although an azimuth symmetry is expected by the Rayleigh's Theory (refs. 31, 32). The plausible explanation of this asymmetry in terms of finite size effects was offered by Theimer (ref. 33). However, the calculations of Feiock (ref. 34) do not support this explanation. For the purpose of further clarification, measurements of scattered radiation were made from argon at backward and forward directions of 54°44' with respect to the incident beam.

It was found that there is no noticeable asymmetry in argon gas. Because of this disagreement, it was decided to perform 54°44' back scattering measurements that might further reduce the finite size effects. The measurements were made for He, H_2, Ar, O_2, and N_2 and were corrected for the changes in incident intensity which were less than 5 per cent. The plots of these measurements are straight lines as predicted by the equation (1) and are shown in Figures 3 through 7. The slopes of these straight lines are the measure of the Rayleigh scattering cross sections. The relative cross sections were derived from the slopes of the lines for 90° and 54°44' scattering. The results are given in Table 1. As shown in the Procedure Section, the cross sections derived from 54°44' scattering are independent of the depolarization ratio of the gas molecules. These results indicate that within the experimental error, the depolarization ratios have no noticeable effect on the scattering cross sections at 6328Å. However, this influence may be significant at shorter wavelengths in vacuum ultraviolet region. If we take into consideration the upper limit $\rho_n = 0.5$ derived by Chandrasekhar (ref. 30), then the maximum error caused by the depolarization ratio would be about 20 per cent (ref. 16, also see Appendix A). In the table are also given similar measurements at 6943 and 1215.7Å obtained by other investigators. The comparison of these results shows that the values of cross sections obtained at 6943Å by Rudder and Bach are about 25 per cent lower than our measured values at 6328Å. The 5 per cent error in our measurements cannot account for this discrepancy. However, when the values of scattering cross sections in the visible region (values at 6943 and 6328Å) are compared with similar values measured in the vacuum ultraviolet at 1215.7Å, the discrepancy becomes much larger. This discrepancy in the results at these wavelengths cannot be explained in terms of either the depolarization ratios or the sources of experimental error. Therefore, in order to resolve this problem, an integrated laboratory study is required to carry out such measurements as a function of wavelength from the visible to ultraviolet region of the electromagnetic spectrum.

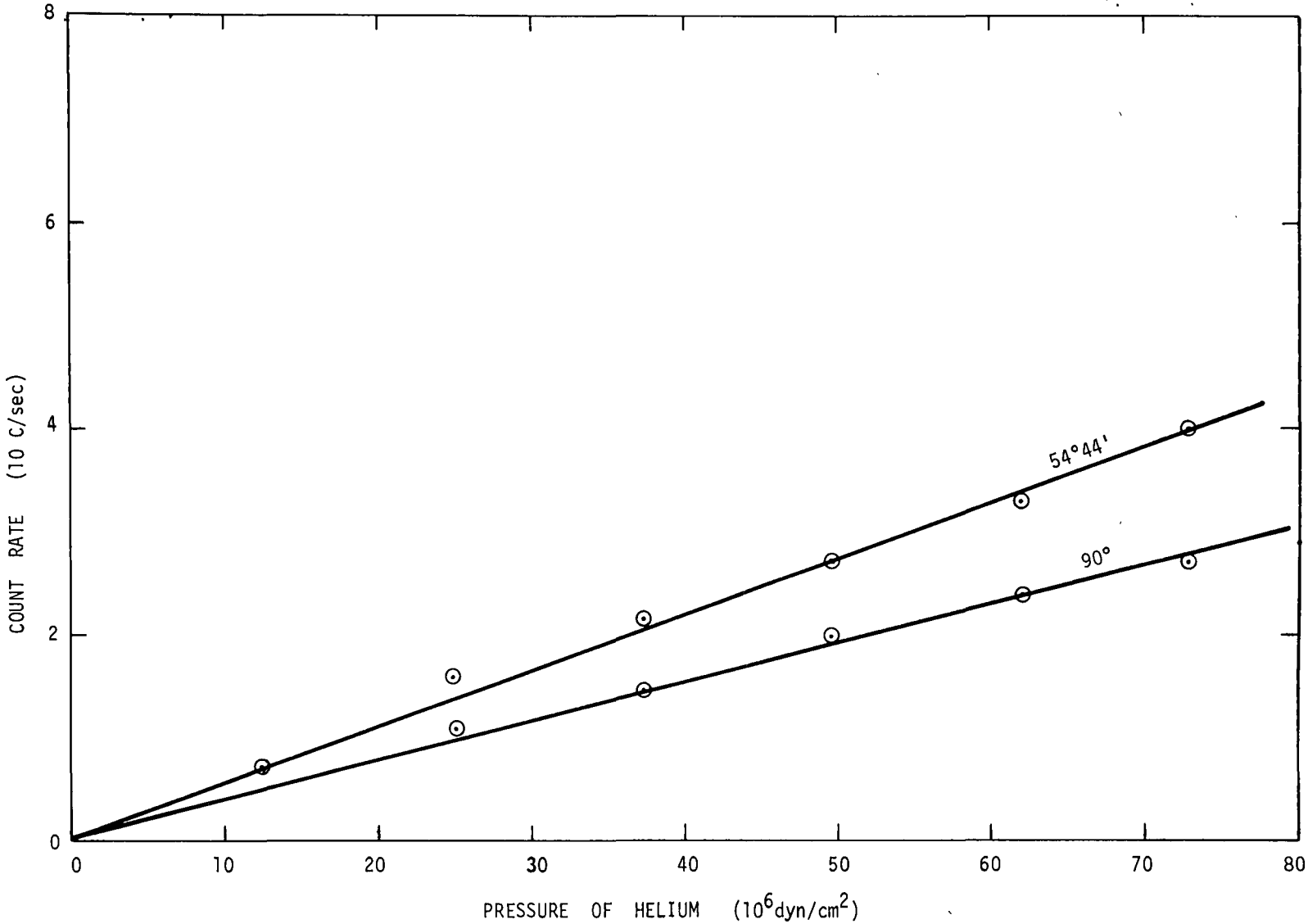

Figure 3. Photomultiplier response from 90° and 54°44' scattering of 6328Å radiation for helium as a function of pressure.

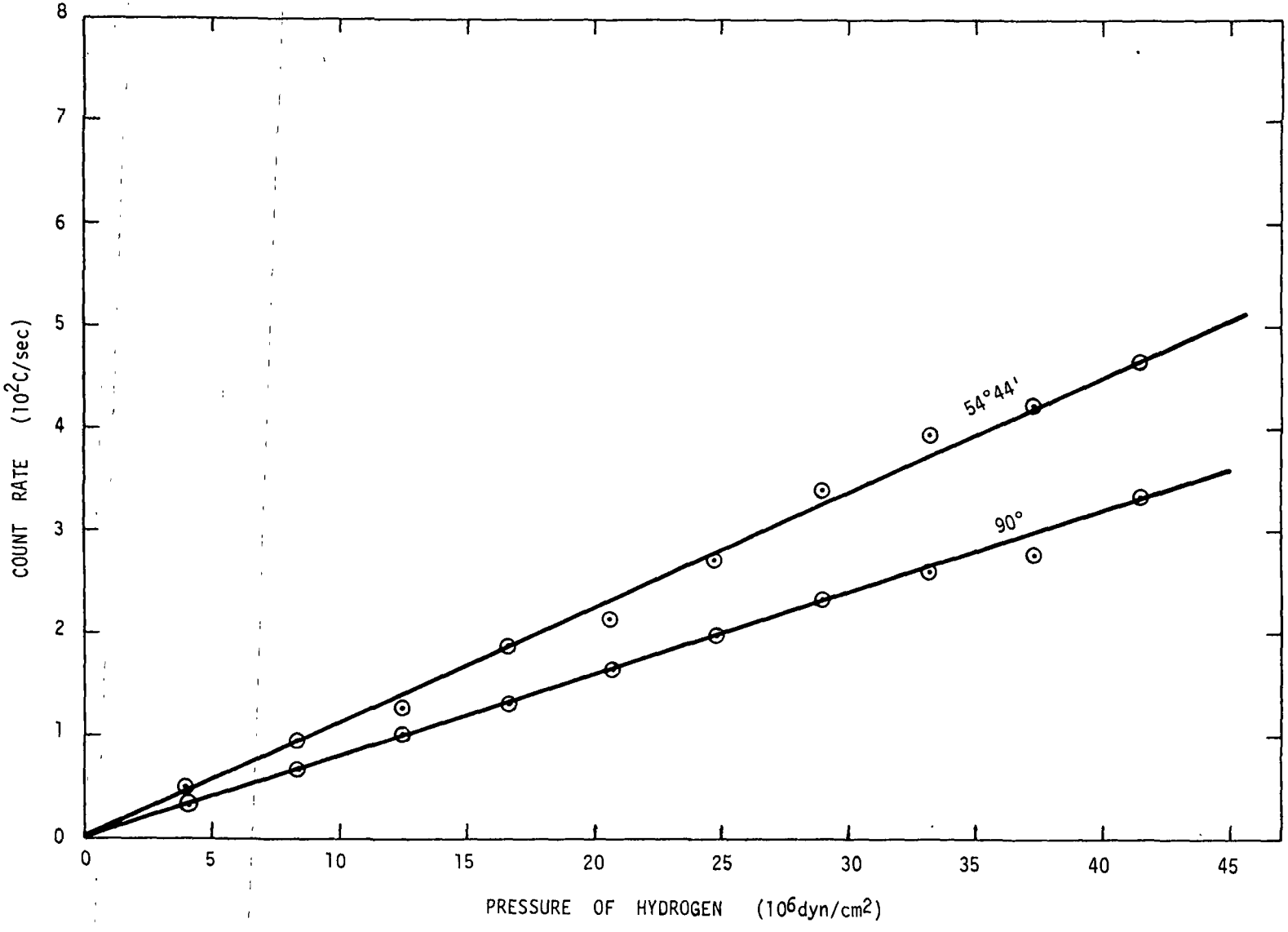

Figure 4. Photomultiplier response from 90° and 54°44' scattering of 6328Å radiation for hydrogen as a function of pressure.

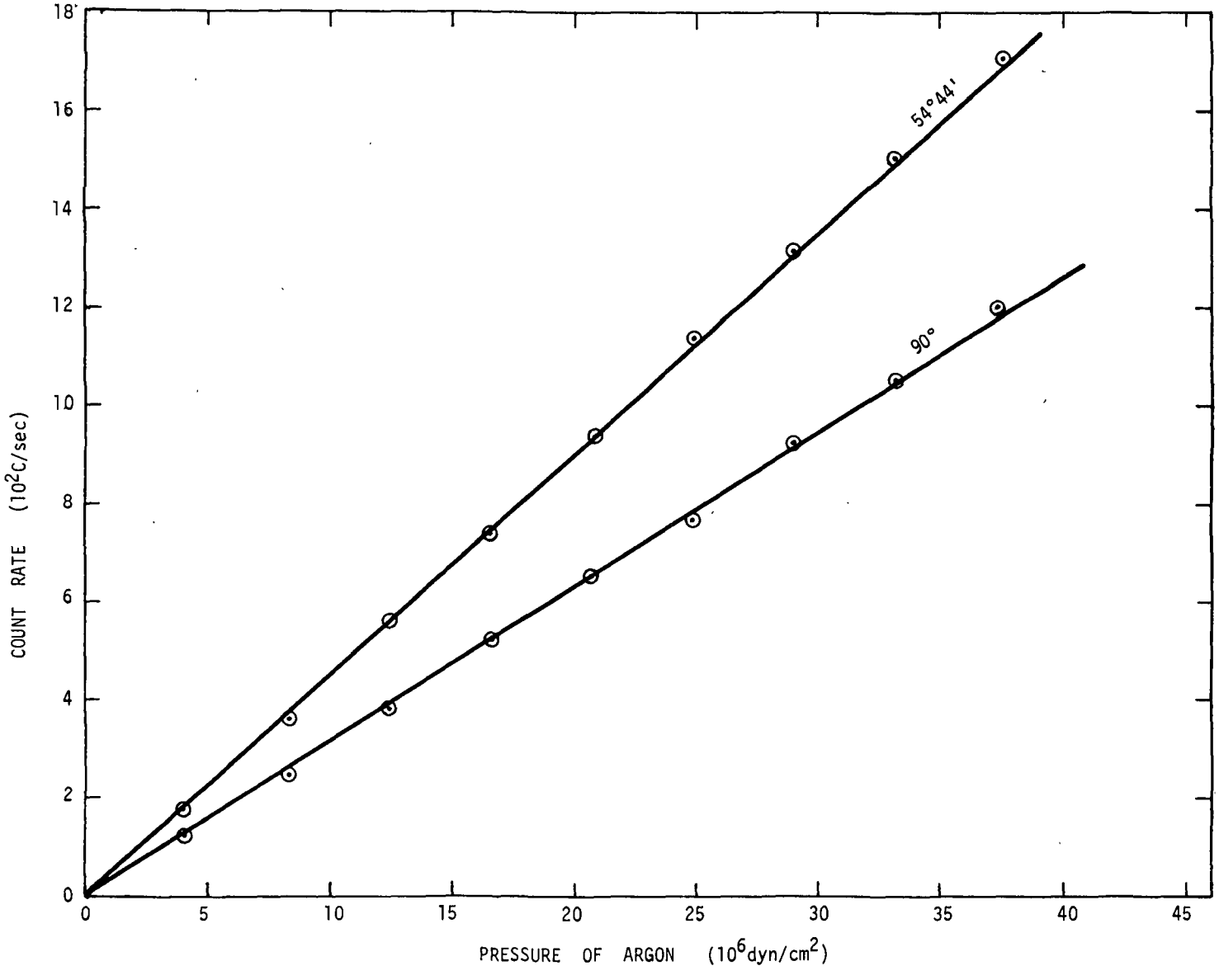

Figure 5. Photomultiplier response from 90° and 54°44' scattering of 6328Å radiation for Argon as a function of pressure.

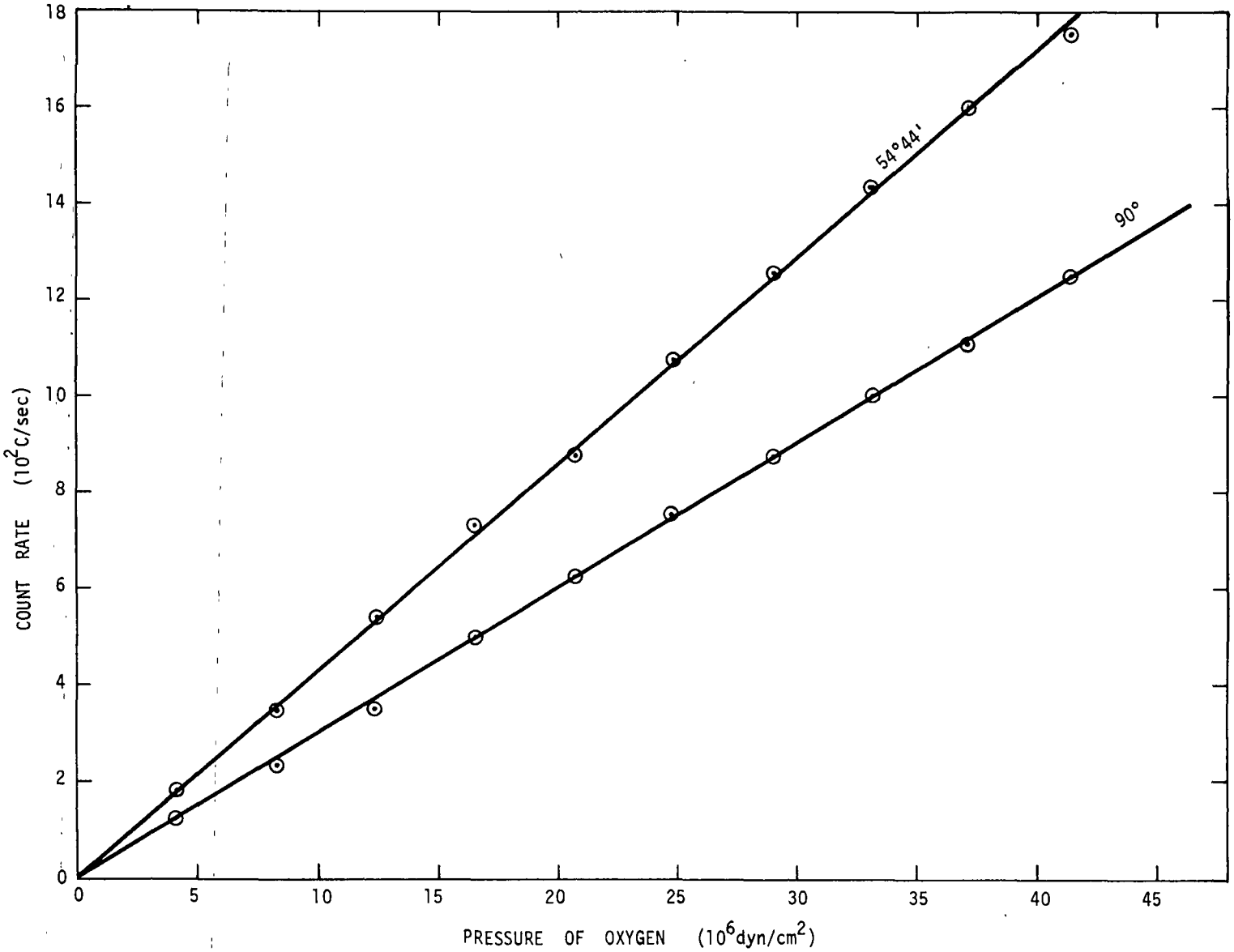

Figure 6. Photomultiplier response from 90° and 54°44' scattering of 6328Å radiation for oxygen as a function of pressure.

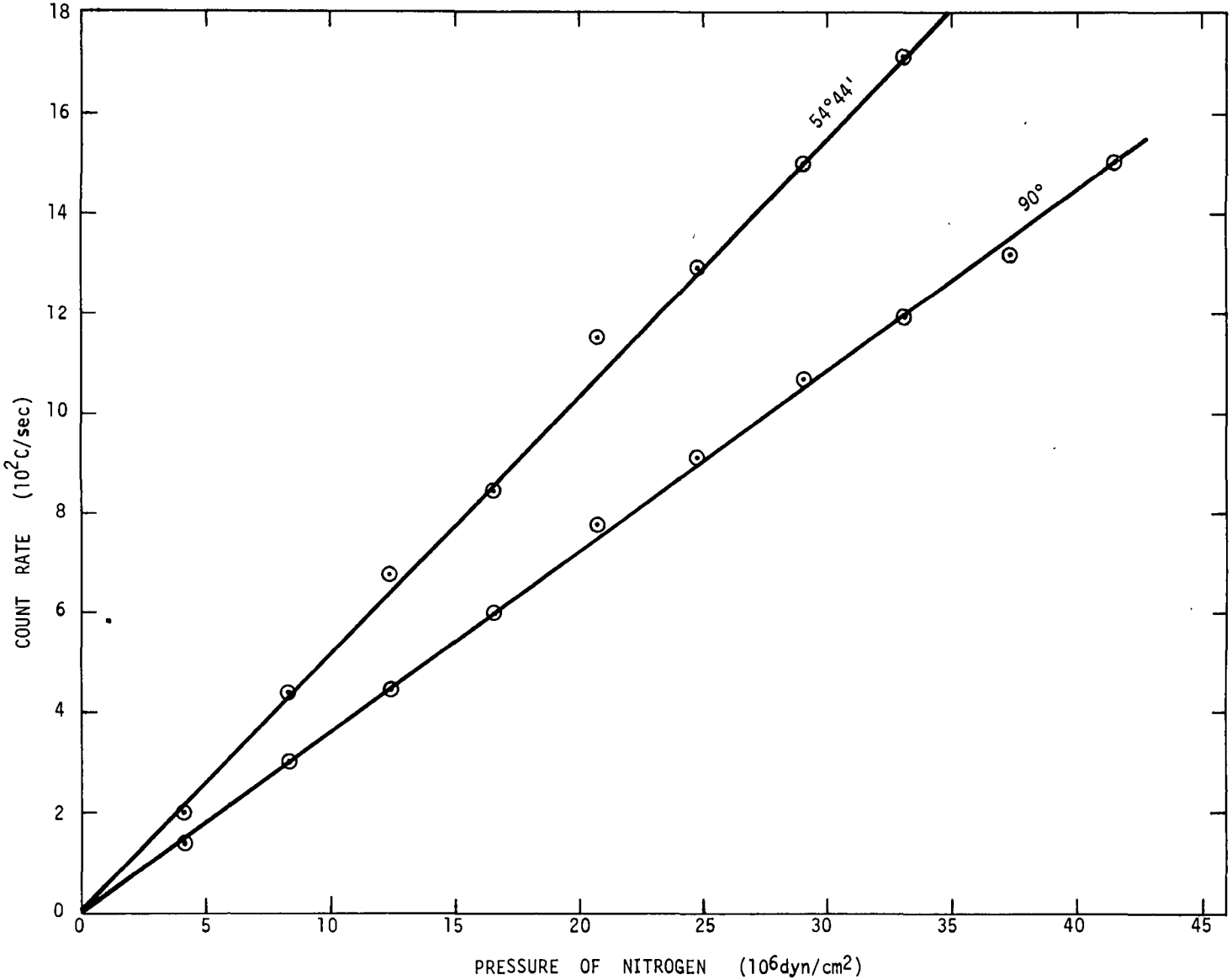

Figure 7. Photomultiplier response from 90° and 54°44' scattering of 6328Å radiation for nitrogen as a function of gas pressure.

TABLE 1. RELATIVE RAYLEIGH SCATTERING CROSS SECTIONS

FOR SEVERAL GASES

GAS	HELIUM	HYDROGEN	ARGON	OXYGEN	NITROGEN
90° Scattering at 6328Å (measured)	1.00	20.94	82.56	78.97	94.89
54°44' Scattering at 5328Å (measured)	1.00	20.68	82.72	79.04	94.98
60° Scattering at 6943Å (ref. 26)	1.00	14.80	63.52	--	71.62
90° Scattering at 1215.7Å (ref. 16)	1.00	63.64	181.80	--	200.00
90° Scattering at 1215.7Å (ref. 15)	--	60.00	181.80	--	200.00

APPENDIX A

Scattering Cross Sections

For this purpose let us examine the equations (2) and (3) which show that the measurements of Φ_{54} and Φ_{90} as a function of pressure will provide straight lines, the slopes of which are the measures of the scattering cross section. If σ_s^i and σ_s^j denote the Rayleigh scattering cross sections of i and j molecules, then it may be shown from equation (2)

$$\frac{\sigma_s^i}{\sigma_s^j} = \frac{(\text{slope})_{54}^i}{(\text{slope})_{54}^j} , \qquad \text{(a)}$$

and from equation (3)

$$\frac{\sigma_s^i}{\sigma_s^j} = \frac{(\text{slope})_{90}^i}{(\text{slope})_{90}^j} \; \frac{(2 + \rho_n^i)}{(1 + \rho_n^i)} \; \frac{(1 + \rho_n^j)}{(2 + \rho_n^j)} . \qquad \text{(b)}$$

If we assume that the relative cross section of the nitrogen molecule with $\rho_n = 0.0214$ is determined with respect to another gas molecule with $\rho_n = 0$, then

$$\frac{\sigma_s^{N_2}}{\sigma_s^j} = \frac{(\text{slope})_{54}^{N_2}}{(\text{slope})_{54}^j} = 0.99 \, \frac{(\text{slope})_{90}^{N_2}}{(\text{slope})_{90}^j} . \qquad \text{(c)}$$

Therefore, from equation (c) it may be concluded that in the visible region where ρ_n values for most gas molecules are very small, the effect of normal depolarization ratios on the scattering cross sections will be negligible as compared to the experimental error. However, the influence of ρ_n may be significant in the vacuum ultraviolet where large values of ρ_n are expected. With an upper limit of $\rho_n = 0.5$, the maximum error caused by the normal depolarization factor would be about 20 per cent. Therefore, this technique might have some advantage in the vacuum ultraviolet region.

APPENDIX B

Depolarization Ratios

The equation (4) in the Procedure Section of the text is

$$\frac{\Phi_{54}}{\Phi_{90}} = \frac{2}{3} \frac{V_{54}}{V_{90}} \left(\frac{2 + \rho_n}{1 + \rho_n}\right) \qquad (d)$$

$$= \frac{2}{3} \frac{V_{54}}{V_{90}} (2 + \rho_n)(1 - \rho_n)$$

$$= \frac{2}{3} \frac{V_{54}}{V_{90}} (2 - \rho_n - \rho_n^2) \qquad (e)$$

Since ρ_n is very small compared to unity, ρ_n^2 may be neglected. Therefore, to first order approximation

$$\frac{\Phi_{54}}{\Phi_{90}} = \frac{2}{3} \frac{V_{54}}{V_{90}} (2 - \rho_n) \qquad (f)$$

$$\frac{d}{d\rho_n} \left(\frac{\Phi_{54}}{\Phi_{90}}\right) = -\frac{2}{3} \frac{V_{54}}{V_{90}} = \text{constant} \quad . \qquad (g)$$

From equation (g), it is clear that the variations in Φ_{54}/Φ_{90} are independent of ρ_n. Therefore, it may be concluded that small variation in $\frac{\Phi_{54}}{\Phi_{90}}$ will require large variation in the value of ρ_n in order to satisfy the equations (e) and (f). For the purpose of error analyses, we can obtain the following expression using the equation (5) in the text.

$$\frac{\Delta\rho_n}{\rho_n} \approx 100 \frac{\Delta(\Phi_{54}/\Phi_{90})}{(\Phi_{54}/\Phi_{90})} \quad , \qquad (h)$$

for $\rho_n = .0214$. This expression (h) is approximately valid for most of the gases in the visible region where the values of ρ_n are quite small. However, with an upper limit of $\rho_n = 0.5$, the above expression becomes

$$\frac{\Delta\rho_n}{\rho_n} \approx 10 \frac{\Delta(\Phi_{54}/\Phi_{90})}{(\Phi_{54}/\Phi_{90})} \quad . \qquad \text{(i)}$$

It is obvious from the expressions (h) and (i) that the error in the ρ_n values would be about 10 to 100 times more than the experimental error in the intensity measurements.

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