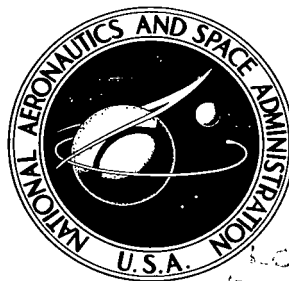


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ABSOLUTE RAYLEIGH SCATTERING
CROSS SECTIONS OF GASES AND
FREONS OF STRATOSPHERIC INTEREST
IN THE VISIBLE AND ULTRAVIOLET REGIONS

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| 16. Abstract The laboratory measurements of absolute Rayleigh scattering cross sections as a function of wavelength are reported for gas molecules He, Ne, Ar, N ₂ , H ₂ , O ₂ , CO ₂ , CH ₄ and for vapors of most commonly used freons CCl ₂ F ₂ , CBrF ₃ , CF ₄ , and CHClF ₂ . These cross sections are determined from the measurements of photon scattering at an angle of 54°44' which yield the absolute values independent of the value of normal depolarization ratios. The present results show that in the spectral range 6943-3638Å, the values of the Rayleigh scattering cross section can be extrapolated from one wavelength to the other using 1/λ ⁴ law without knowing the values of the polarizabilities. However, such an extrapolation can not be done in the region of shorter wavelengths. | | | |
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ABSOLUTE RAYLEIGH SCATTERING CROSS SECTIONS OF GASES AND FREONS OF
STRATOSPHERIC INTEREST IN THE VISIBLE AND ULTRAVIOLET REGIONS

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INTRODUCTION

Absolute Rayleigh scattering cross sections provide useful data for defining the magnitude of photon molecule interactions. Generally, these cross sections are computed from the index of refraction at a wavelength and then extrapolated to other wavelengths using $1/\lambda^4$ law (refs. 1 and 2). It would be appropriate to remark that the refractive indices for gases tabulated in most handbooks are at 5893\AA , Na D lines. In recent years, however, there are a few publications on the indices of refraction at other wavelengths (see bibliography). The earlier measurements (refs. 3 thru 15) on the Rayleigh scattering using conventional light sources were limited in scope because of the experimental difficulties concerning the intensity of light source, bandwidth, collimation, and above all the lack of instrumentation for detecting the weak scattered signals. In recent years, these difficulties have been overcome with the availability of lasers and with the improved technology in light detection. Several investigators (refs. 16 thru 30) have used these recent techniques for measuring the photon scattering from atoms and molecules at selected laser wavelengths in the visible region. However, there are only a few measurements in the vacuum ultraviolet (refs. 31 thru 35).

To the best of our knowledge, there is no publication concerning the direct measurements of the absolute Rayleigh scattering cross sections for atmospheric gas molecules as a function of wavelength. For the present investigation, we have employed an Ar-ion laser (Model CR-5) for the wavelengths 5145, 4880, 4579, and 3638\AA and a He-Ne laser (Spectra Physics Model 132) for the wavelength 6328\AA . The polarization of the radiation emitted from Ar-ion laser is found to be 95% in the the direction perpendicular to the scattering plane. The He-Ne laser is linearly polarized in the same direction. This study is to

report for the first time such measurements for He, Ne, Ar, N₂, H₂, O₂, CO₂, CH₄ as a function of wavelength. In addition, recognizing the great importance of freon vapors as pollutants for problems related to stratospheric studies, similar measurements are also presented for commonly used freons CCl₂F₂, CBrF₃, CF₄, and CHClF₂. The measured cross sections are compared with other measured and calculated data.

THEORETICAL BACKGROUND

The intensity, $I_S(\theta, \eta, \psi)$, scattered by gas molecules of number density, N , per unit solid angle in the direction θ, η , and ψ (fig. 1) from a partially polarized incident beam of intensity, I_0 , whose K vector is along the Y -axis is given by the expression (ref. 35)

$$I_S(\theta, \eta, \psi) = I_0 \sigma_S \frac{3NV}{4\pi} \frac{1}{2+\rho_n} \left[1 + \frac{\rho_n^{-1}}{1+g} \cos^2\theta + g \frac{\rho_n^{-1}}{1+g} \cos^2\psi \right], \quad (1)$$

where V is the scattering volume; σ_S is the total cross section for Rayleigh scattering; $\rho_n = 6\gamma^2/(45\alpha_0^2 + 7\gamma^2)$ is the normal depolarization factor and is zero when γ representing the anisotropic property of the gas molecule is zero. The quantity α_0 represents the mean polarizability of the molecule. The factor $g = (E_x/E_z)^2$ is related to the degree of polarization, P , of the incident beam by the relation $P = (1-g)/(1+g)$. It should be pointed out that when $\theta = \eta = \psi = 54^\circ 44'$, the scattered I_S is equal to the average intensity of the scattered radiant energy and is independent of ρ_n and g . However, if I_S is observed for conditions such that $\theta = \psi$, then I_S is independent of g only.

Utilizing the direction cosine relation, namely, $\cos^2\theta + \cos^2\eta + \cos^2\psi = 1$, the equation (1) can be written as

$$I_S(\theta, \eta, \psi) = I_0 \sigma_S \frac{3NV}{4\pi} \frac{1}{2+\rho_n} \left[1 + \frac{\rho_n^{-1}}{1+g} (1 - \cos^2\eta) + \frac{(\rho_n^{-1})(g-1)}{1+g} \cos^2\psi \right], \quad (2)$$

where η represents the direction of observation of the scattered radiation with respect to the direction of incident beam. In the present experimental setup, an incident beam of partially polarized radiant energy travels along the Y -axis (fig. 1) with electric vectors E_x and E_z . Also, in this experiment $\psi = 90^\circ$ and $\eta = 54^\circ 44'$. Substituting these values, the expression (2) may be written as

$$I_S(\theta, \eta, \psi) = I_0 \sigma_S \frac{NV_{54}}{4\pi} \left[\frac{1}{2+\rho_n} \frac{1+3g+2\rho_n}{1+g} \right], \quad (3)$$

where V_{54} is the scattering volume when the scattered radiation is observed at $\eta = 54^\circ 44'$. In the visible region of electromagnetic spectrum, the values of ρ_n for most gases are

very small [ref. 36; $\rho_n(N_2) = 0.0214$; $\rho_n(O_2) = 0.0565$; $\rho_n(H_2) = 0.0188$]. In such cases $\rho_n \approx 0$, and the expression (3) for the partially polarized incident beam becomes

$$I_S(\theta, \eta, \psi) = I_0 \sigma_s \frac{\sqrt{54}}{4\pi} \frac{3g + 1}{2(1 + g)} N. \quad (4)$$

It should be pointed out that in vacuum ultraviolet region ρ_n may not be treated as insignificant.

The following two cases of special interest often arise and may easily be derived from equation (3).

1. Case I. When the incident beam of radiant energy is unpolarized. In this case $E_x = E_z$ and, therefore, $g = 1$. Thus, the expression for the intensity of radiation scattered from an unpolarized incident beam becomes (refs. 30, 31, 36)

$$I_S(\theta, \eta, \psi) = I_0 \sigma_s \frac{\sqrt{54}}{4\pi} N. \quad (5)$$

Here, it is important to note that I_S is independent of ρ_n . Therefore, the equation (5) can be used in the entire spectral region ranging from visible to the vacuum ultraviolet without knowing the specific value of ρ_n .

2. Case II. When the incident beam of radiant energy is plane polarized in the direction perpendicular to the scattering plane. In this case $E_x = 0$ and, therefore, $g = 0$. Thus, the expression for the intensity of radiation scattered from a linearly polarized incident beam becomes

$$I_S(\theta, \eta, \psi) = I_0 \sigma_s \frac{\sqrt{54}}{4\pi} N \frac{2\rho_n + 1}{2 + \rho_n} \quad (6)$$

Unlike the Case I, it should be emphasized that in the case of polarized incident radiation, the scattered radiation I_S is not independent of ρ_n . This means that the specific values of ρ_n need to be known in the spectral region where the contribution of normal depolarization to the scattered radiation is significant. However, in the visible, when $\rho_n \approx 0$, the equation (6) may be written as

$$I_S(\theta, \eta, \psi) = \frac{1}{2} \left[I_0 \sigma_s \frac{\sqrt{54}}{4\pi} \right] N \quad (7)$$

It should be noted that the equations (5) and (7) are identical except for the factor 1/2 in the right hand side of equation (7).

EXPERIMENTAL ARRANGEMENT

A block-diagram of the experimental arrangement is shown in Figure 2. It employs a laser as a light source, a gas pressure vessel called a scattering chamber for filling the samples of gases, and the photon counting equipment for monitoring the scattered and incident light signals. The scattering chamber has been fabricated with great precision which almost eliminated the generally encountered sources of error in scattering measurements. Briefly, it is made of stainless steel and can withstand pressures up to 100 atm. There are seven ports in the chamber sealed with sapphire windows. The location of the ports is as shown in the figure. The laser beam is expanded and collimated by using a suitable beam expander-cum-collimator before entering the incident port. The beam is further collimated and made into a rectangular cross section (19 mm x 5 mm) by means of a rectangular slit (S) attached to the incident port (1). The ports numbering 2 through 6 are provided with collimators each fitted with 3 rectangular aperture slits (19 mm x 5 mm) in series and are such that the front aperture slit in each collimator is quite close to the scattering volume. There are five light traps (LT) each situated opposite to the ports (2 through 6) which hold the photomultiplier for measuring the light signals scattered at 27° , $54^\circ 44'$, 90° , $125^\circ 16'$, and 153° . These angles are measured with respect to the direction of incident beam. The configuration of the light traps is shown in the left hand corner of the figure and is similar to that used by Pritchard and Elliott (ref. 38). The light traps are made of stainless steel and inside surfaces hold the polished plates of Corning black glass which act as the reflecting surfaces. Most of the light entering the trap is absorbed at the first glass plate. The specular reflection is directed further into the trap, where it is absorbed during additional reflections. The light leaving the trap is from non-specular reflection at the first surface. The glass used in these traps has a low non-specular reflectance of about 10^{-6} (ref. 38). In the center of the scattering Chamber, there is a provision for mounting a calibration carriage and a front surface mirror which is used in measuring the angle of light scattering and in aligning the rectangular apertures in the light collimators, (C). The inside surface of the chamber, the light traps, and the collimators are coated with flat-black-chrome-finish that provides the dark background which in conjunction with the light traps minimize any spurious radiation reaching the detector. The performance of the inside system was such that the chamber under vacuum and laser light-on provided a background signal which was a few percent of scattered signal under all pressure conditions in the chamber. There are other ports, not shown in the figure, which are used for gas outlet, pumping, safety valve, and pressure gauge.

The light sources used in the present measurements are an Ar-ion laser (Coherent Radiation Model CR-5) for wavelengths at 5145, 4880, 4579, and 3638Å and a He-Ne laser

(Spectra Physics Model 132) at $6328\overset{\circ}{\text{Å}}$. The output of the laser in use is monitored constantly for changes in radiation flux by means of a photomultiplier (PM2; EMI 9558Q). The intensity of the laser beam was reduced by inserting a series of four neutral density filters in the path of beam before entering the photomultiplier housing. The attenuation factors of the neutral density filters were premeasured. The use of the 45° mirror was to divert the laser beam. The output of the Photomultiplier tube (PM2) in conjunction with an amplifier-discriminator (AD2; SSR 1120) and data converter is used as an external clock input of the digital synchronous computer (SSR 1110). This output is also recorded on a strip chart recorder (HP Model 7101B) and thus provides the monitoring of the changes in the incident radiation. The photon counting technique is used for the measurements of both the incident and scattered radiation. For the purpose of measuring scattered radiation, another photomultiplier tube, (PM1; EMI 9558Q), enclosed in a housing (SSR 1151), an amplifier-discriminator (AD1; SSR 1120), a digital synchronous computer (SSR 1110), and a digital recorder (HP Model 5055A) modified to match the SSR photon counting system, are used. The high pressure in the scattering chamber and a defocussing magnetic assembly having an opening of 19 mm in diameter in front of the photomultiplier tube are used to enhance the signal-to-noise ratio which is an essential requirement for measuring the weak scattered light signals. The use of the defocussing magnetic assembly alone improved the signal-to-noise ratio by about a factor of 10 in the present setup. The possible changes in the response of the photomultiplier are intermittently checked by means of a neon reference source (N) which was employed with the scattered intensity monitoring tube (PM1). Here, it should be pointed out that no change in the response of the tube was observed during the period of observations. Besides, a mechanical chopper (PAR Model 125) is employed in front of this photomultiplier housing to account for the changes in the dark counts of the photomultiplier.

OBSERVATIONS AND RESULTS

The examination of general expression (1) for the scattered intensity shows that the values of I_s/I_0 are linearly proportional to the number density of the scatters. The plot of I_s/I_0 vs pressure of gas [$P = P_0 (N/N_0) (T/T_0)$] is thus a straight line which will yield the average value of Rayleigh scattering cross section from its slope. Here p is a pressure corresponding to number density N and temperature T ; N_0 is a Loschmidt number and P_0 and T_0 are standard pressure and temperature, respectively. Therefore, for the determination of the absolute value of Rayleigh scattering cross section it is required to measure the values of I_s/I_0 and the scattering volume V .

Measurement of Intensity Ratio I_S/I_0

Let us assume that I_0 is uniformly distributed radiation flux incident on the scatters in the scattering volume V and I_S is the radiation scattered into a solid angle defined by the scattering volume and other geometrical parameters of the experiment. Since the response of the detector is proportional to the radiation falling on the photocathode, the expression

$$I_S/I_0 = C_S/C_0 \quad (8)$$

is satisfied provided the same detector is used for I_S and I_0 . Here, C_S and C_0 are the count rates corresponding to I_S and I_0 , respectively. This means that the problem of measuring the value of I_S/I_0 is to measure the responses of the detector corresponding to I_S and I_0 . For this purpose, the photomultiplier tube PM1 with an applied voltage of 900V (this voltage was kept constant throughout the experiment) was used at the exit port number 7 of the scattering chamber. A set of four neutral density filters of known attenuation factors were inserted in the beam of the laser light. These filters reduced the intensity of radiation falling on the photomultiplier by about 10^{-8} . The multiple reflections arising from these filters were considered. Their contribution was calculated to be 0.01% and thus not taken into account. From the known values of transmission factor of sapphire window (the transmission factors of the sapphire windows were premeasured - see Appendix A) and the attenuation factors of the neutral density filters, the value of C_0 was determined for a particular wavelength. The error estimated in the determination of C_S/C_0 was about $\pm 9.3\%$ (Appendix A). The same procedure was repeated for other wavelengths. At this stage onward, the photomultiplier PM1 with the necessary equipment (amplifier-discriminator AD1, synchronous computer, and digital printer) was used for the measurement of scattered radiation. However, to monitor the fluctuations in the laser output and subsequently to account for this fluctuation second photomultiplier PM2 with another amplifier-discriminator AD2 and data converter was used. The necessary setup to accomplish this is shown in Figure 2. The output from the data converter was recorded on the strip chart recorder for monitoring and initial setting of I_0 to the same value for each run of the observation. Also, this output was fed as an input to the external clock of the synchronous computer and thus the fluctuations in incident intensity were accounted for.

Measurement of Scattering Volume

The measurement of scattering volume is the most tedious job. Therefore, the experiment was planned and set up in such a way that the incident light beam has almost no divergence in the domain of the scattering volume. The attempt was also made to minimize

the solid angle subtended by the scattering volume at a point in the center of photocathode of the tube. For this purpose a series of rectangular apertures were so arranged that one of the aperture was very close to the scattering volume in every case of scattering angle of observation. If the solid angle so subtended is zero, the scattering volume viewed at an angle η will vary with $1/\sin \eta$ with respect to the scattering volume at 90° , namely,

$$V_{90}/V_{\eta} = \sin \eta \quad (9)$$

where, V_{90} and V_{η} are the scattering volumes viewed by the photomultiplier at the scattering angles of observation 90° and η , respectively. For a specific angle, $54^\circ 44'$, of observation it has been shown (ref. 30) that

$$V_{54^\circ 44'} = 1.225 V_{90} \quad (10)$$

The detailed discussion of equations (9) and (10) is given in reference (30). As pointed out earlier that in our experiment the incident and scattered beams are almost collimated. In such a geometry the scattering volume defined by the perpendicular intersection of these beams (scattering observed at 90°) would be a rectangular parallelepiped whose volume can be determined by measuring its three sides. In order to measure the dimensions of the rectangular parallelepiped, a circular aperture of 50 micron size was placed in front of the photomultiplier (PM1). This aperture can be moved along the X and Y axes by means of a XY micrometer translation stage. The aperture is scanned along the X-axis at a fixed Y-position. This is recorded on a strip chart recorder as a trapezoidal function with some diffraction pattern superimposed on the top portion. The width of the function at half maximum points is taken as the width at that particular Y-position. The aperture is moved 1 mm along the Y-axis and the width is measured again. This procedure is repeated for every 1 mm interval along the Y-axis. The average of all these values is taken as the width of the parallelepiped along the X-axis. Similar measurements are taken along the Y-axis (keeping fixed position along X-axis) and the height determined. The depth of the rectangular parallelepiped is then determined by moving the aperture and the detector assembly to the reference port (port no. 7) of the scattering chamber and repeating the procedure along the Z-axis. The product of these three measured quantities is taken to be the scattering volume whose value was found to be $V_{90} = 0.64 \text{ cm}^3$. Having determined the value of V_{90} , the circular aperture is then removed from the view of the detector. In order to determine the value of V_{54} , the procedure adopted is as follows.

The scattering chamber is pressurized to a pressure of about 35 atm and the scattered radiation is observed at 90° and $54^\circ 44'$. It is found that

$$I_s (54^\circ 44') = 1.22 I_s (90^\circ)$$

In the spectral bandpass at 4880\AA where the above intensities are measured the contribution of normal depolarization factor may be assumed to be negligible (ref. 30). Therefore, the scattering intensities may be taken as proportional to the respective scattering volumes,

$$\frac{I_s (54^\circ 44')}{I_s (90^\circ)} = \frac{V_{54}}{V_{90}} = 1.22$$

It should be pointed out that if the incident and scattered beams were perfectly collimated, the ratio V_{54}/V_{90} would have been 1.225. However, there was a small solid angle, $\Omega = 4.35 \times 10^{-4}$ Sr, subtended by the scattering volume at the detector. The error estimated in the determination of scattering volume is about $\pm 5\%$.

Scattering Cross Section Measurement

The sample gases supplied by Air Products and Chemicals, Inc., with percentage purity of He - 99.995, Ne - 99.99, Ar - 99.998, N_2 - 99.998, O_2 - 99.99, H_2 - 99.997, CO_2 - 99.9, and CH_4 - 99.7 were used in the present measurements. Also because of the great interest in the scattering properties of freons as pollutants which might not only cause the depletion of stratospheric ozone but might also affect the atmospheric albedo, the Rayleigh scattering cross sections of most commonly used freons (12 - CCl_2F_2 - 99%; 13B1 - $CBrF_3$ - 99%; 14 - CF_4 - 99.7%; and 22 - $CHClF_2$ - 99%) supplied by Matheson were measured. All these samples of gases and freon vapors were checked for the attenuation of radiation at 6328, 5145, 4880, 4579 and 3638\AA . It was found that there was no measurable attenuation at these wavelengths for these molecules with the exception of CH_4 which showed an attenuation compatible to the attenuation cross section of $2.5 \times 10^{-23} \text{ cm}^2$ at 3638\AA . However, it may be pointed out that the attenuation of radiation by absorbing species, if present, could be accounted for as the pathlengths from the scattering volume to the detectors were equivalent in the present setup (ref. 33). This was accomplished in the case of CH_4 . The samples of gases and vapors were allowed to pass through a series of millipore filters (VSWP 047 00) which filtered the dust particles of size larger than 25 μ . The use of these filters minimized the scattering due to dust particles which are believed to have high scattering cross section as compared to the Rayleigh scattering. The filtered gas and vapor samples were then allowed to enter into the scattering chamber. The gas pressures were measured by a Heiss pressure gauge which was calibrated in absolute terms in the range of 0 - 100 atm. First, the scattered signals were measured as a function of scattering angles, namely, 27° , $54^\circ 44'$, 90° , $125^\circ 16'$, and 153° (measured with respect to

the direction of incident beam) in order to check the asymmetry in the scattering. For this purpose, the scattering chamber was pressurized to 34 atm by oxygen gas. It was found that there is no noticeable asymmetry in oxygen gas (see Figure 3). It should be pointed out that the dust particles of size smaller than 25 μ would introduce + 2.2% disymmetry which is small as compared to the error in the measurements (Appendix B). It would be appropriate to remark that George et al (refs. 18 and 19) noted a forward enhanced asymmetry in the observations for argon and xenon, although an azimuth symmetry is expected from the Rayleigh's theory (refs. 39 and 40). A plausible explanation of this asymmetry observed by George et al was offered by Theimer (ref. 41) in terms of finite size effects. However, the calculations of Feiock (ref. 41) do not support this explanation. In spite of the fact that we did not notice the enhanced asymmetry in forward direction, it was decided to perform back scattering measurements. This would minimize the controversy of the effects of dust particles in the present measurements. In addition, these measurements would be independent of normal depolarization factor, ρ_n , if the measurements are performed at a scattering angle $54^\circ 44'$. The data presented below are for backscattering at $54^\circ 44'$.

The measurements of scattered signal were made as a function of pressure for various gases He, Ar, N₂, and O₂ at wavelengths 3638, 4579, 4880, 5145 $\overset{\circ}{\text{A}}$ emitted by Ar-ion laser and at 6328 $\overset{\circ}{\text{A}}$ emitted by He-Ne laser. The plots of these measurements $I_s(54^\circ)/I_0$ vs gas pressure are straight lines as predicted by the equation (1) and are shown in Figures 4 thru 7. Additional, similar measurements were performed for Ne, H₂, CO₂, CH₄, and most commonly used freons F-12, F-13B1, F-14, and F-22 at 3638 and 5145 $\overset{\circ}{\text{A}}$ wavelengths. For reference, similar measurements were repeated for O₂. These measurements are shown in Figures 8 and 9. Since the relative measurements in the wavelength range 3638-6328 $\overset{\circ}{\text{A}}$ were found to be the same, the measurements of scattered radiation at other wavelengths (4579, 4880, and 6328 $\overset{\circ}{\text{A}}$) were not made. In our determination of scattering cross section, the equation (4) for partially polarized radiation and the equation (7) for linearly polarized radiation were employed. Obviously, the slopes of the straight line plots are the measures of the Rayleigh scattering cross sections. First, the values of the slopes for He, Ar, N₂ and O₂ were determined (Figures 4 thru 7) and then utilizing the measured values of

$$I_s(\lambda, 54^\circ)/I_0(\lambda) = C_s(\lambda, 54^\circ)/C_0(\lambda)$$

and the scattering volume V_{54} , the absolute values of the Rayleigh scattering cross sections were determined and are shown in Table I. The values of the relative cross sections were determined at 3638 and 5145 $\overset{\circ}{\text{A}}$ for Ne, H₂, CO₂, CH₄, and freons and normalized with respect to the values of O₂. Knowing the absolute values of the Rayleigh scattering cross section of O₂ at these two wavelengths, the relative measurements of Ne, H₂, CO₂, CH₄, and freons were converted into absolute units. The values thus obtained are also shown in Table I. The values for these molecules at other wavelengths (4579, 4880, and 6328 $\overset{\circ}{\text{A}}$)

were determined using λ^{-4} law. The estimated error in the measured scattering cross section is about $\pm 11.0\%$. In the table (column 7) are also shown the scattering cross section values at Lyman- α line (1215.7Å) measured by Shardanand and Mikawa (ref. 33) and Heddle (ref. 31). In column 8 are given the recent values at Lyman- α measured by Chopra and Heddle (ref. 35). In columns 9 and 10 are the scattering cross sections at 6328 and 3638Å determined by λ^{-4} law extrapolation from the values given in column 4 of Table II. In the last column of Table I are shown the theoretical calculated values of Layman- α (1215.7Å). The value for He is due to Chan and Dalgarno (ref. 43); Ne and Ar are calculated from Dalgarno and Kingston's method (ref. 44) and for H₂ from the method of Dalgarno and Williams (ref. 45). These Layman- α values of He, Ne, Ar and H₂ are cited by Shardanand and Mikawa (ref. 33). It should be pointed out that the indices of refraction are compiled in Table II for ready reference (refs. 46 and 47). The refractive indices of O₂, N₂, Ar, Ne, He, H₂, CO₂ and CH₄ are due to conventional methods of refractometry whereas those of freons are determined from the values of Rayleigh scattering cross sections using the relation

$$\sigma_s = \frac{32\pi^3 (\mu-1)^2}{3N_0^2 \lambda^4} \quad (11)$$

Here, μ is the refractive index at STP, λ is the incident wavelength in Angstrom, and $N_0 = 2.69 \times 10^{19} \text{ cm}^{-3}$ is a Loschmidt number. In column 4 (Table II) are given the values of Rayleigh scattering cross sections which are related to the index of refraction by the above relation.

DISCUSSION

As pointed out in the Introduction the Rayleigh scattering cross sections as a function of wavelength have been measured for the first time. However, there are a few recent measurements at individual wavelengths, mainly at 6943 and 6328Å. In order to compare the results obtained by various investigators, it is appropriate to reduce all the data in the same form and to one wavelength. Since other investigations have expressed their results in the form of differential scattering cross section, $\frac{d\sigma}{d\Omega}$, defined by the relation

$$\frac{d\sigma}{d\Omega} = \sigma_0 \frac{1}{2} (1 + \cos^2 \eta),$$

where

$$\sigma_0 = \frac{4\pi^2 (\mu-1)^2}{N_0^2 \lambda^4} \quad (12)$$

we also changed our values to satisfy the relation (12). Then using λ^{-4} law, the present measured values at 3638Å and some other published data have been reduced to 6943Å and are shown in Table III. For comparison, the actual measured values at 6943Å are given in columns 4, 5, and 6. In the last column are given the theoretical values calculated from the index of refraction. It can be seen that there is a good agreement between the values for all the gases at 6943Å obtained by various investigators except by George et al (ref. 19) whose values are about half of the others. This agreement clearly gives a definite proof of the validity of the process of extrapolation of the value of Rayleigh scattering cross section from one wavelength to the other using λ^{-4} law without knowing the specific values of the mean polarizability of the gas molecules in the spectral range 3638 - 6943Å. This is also shown graphically in Figures 10 thru 13. The λ^{-4} law also suggests that the relative cross sections should be the same for all wavelengths. For the sake of convenience and ready reference such relative cross sections are presented in Table IV. It can be seen that in the spectral range 3638 - 6943Å, the relative values for each gas are the same and agree reasonably well with the value determined from the index of refraction at 5893Å. However, it should be noted that the relative cross section values at Lyman- α (1215.7Å) are strikingly higher. This behavior is also shown in Figures 10 thru 13. The value of scattering cross section at Lyman- α (1215.7Å) for O₂ is not known and therefore, not shown in Figure 13. The higher relative values at Lyman- α suggest that the simple process of extrapolation by λ^{-4} law may no longer be valid in the short wavelength region. These higher relative values in the vacuum ultraviolet may well be due to the wavelength dependent values of the polarizabilities which may be much higher in the vacuum ultraviolet. Therefore, to calculate the values of Rayleigh scattering cross sections, the specific values of the polarizability as a function of wavelength should be known.

TABLE 1. ABSOLUTE TOTAL CROSS SECTIONS FOR RAYLEIGH SCATTERING OF SELECTED GASES AND VAPORS

UNITS: 10^{-27} cm^2

| Wavelength Å Molecule | Present Measured Values * | | | | | refs. 31&33 | ref. 35 | Extrapolated from Table 2 | | Theoretical (see text for refs.) |
|---|---------------------------|-------|--------|--------|--------|----------------|------------|------------------------------|--------|-------------------------------------|
| | 6328 | 5145 | 4880 | 4579 | 3638 | 1215.7 | | 7328 | 3638 | 1215.7 |
| O ₂ | 2.06 | 4.88 | 6.50 | 8.39 | 20.03 | | | 2.20 | 20.19 | |
| N ₂ | 2.24 | 5.61 | 7.26 | 10.38 | 23.82 | 7000 | 5370 | 2.65 | 24.26 | |
| Ar | 2.08 | 5.46 | 7.24 | 10.13 | 23.00 | 6400 | 5470 | 2.38 | 21.78 | 6400 |
| Ne | 0.103 | 0.25 | 0.33 | 0.42 | 1.01 | 150 | | 0.128 | 1.17 | 140 |
| He | 0.036 | 0.086 | 0.115 | 0.15 | 0.35 | 35 | | 0.039 | 0.36 | 35 |
| H ₂ | 0.493 | 1.17 | 1.56 | 2.01 | 4.80 | 2200 | 2280 | 0.53 | 4.80 | 2100 |
| CO ₂ | 7.28 | 17.25 | 23.00 | 29.60 | 70.70 | | | 6.22 | 57.00 | |
| CH ₄ | 5.26 | 12.44 | 16.59 | 21.40 | 51.10 | | | 5.92 | 54.20 | |
| FREONS - IMPORTANT TO STRATOSPHERIC POLLUTION STUDY | | | | | | | | | | |
| Freon-12 CCl ₂ F ₂ | 41.23 | 97.56 | 130.18 | 167.80 | 401.00 | | | 31.42 | 287.60 | |
| Freon-13B1 CBrF ₃ | 28.25 | 66.94 | 89.27 | 115.10 | 276.45 | | | | | |
| Freon-14 CF ₄ | 5.62 | 13.30 | 17.74 | 22.83 | 54.62 | | | | | |
| Freon-22 CHClF ₂ | 25.07 | 59.38 | 79.17 | 102.05 | 243.80 | | | | | |

* Estimated error $\leq \pm 11.0\%$.

TABLE 2. INDICES OF REFRACTION AND TOTAL
RAYLEIGH SCATTERING CROSS SECTIONS

| Molecule | Index of Refraction μ | Wavelength (\AA) | σ_s (10^{-28} cm^2) | ref. |
|--|------------------------------|--------------------------------|---|------|
| O_2 | 1.000271 | 5893 | 27.90 | a |
| N_2 | 1.000296 | " | 33.40 | a |
| Ar | 1.000281 | " | 29.91 | a |
| Ne | 1.0000671 | " | 1.71 | b |
| He | 1.000036 | " | 0.49 | a |
| H_2 | 1.000132 | " | 6.62 | a |
| CO_2 | 1.000448 | " | 7.82 | a |
| CH_4 | 1.000444 | " | 74.73 | a |
| CCl_2F_2 (freon-12) | 1.00133 | 5145 | 996.10 | p |
| CCl_2F_2 (freon-12) | 1.001050 | 6328 | 31.40 | c |
| CBrF_3 (freon-13B1) | 1.0011 | 5145 | 682.8 | p |
| CF_4 (freon-14) | 1.000475 | 5145 | 135.72 | p |
| CHClF_2 (freon-22) | 1.0010036 | 5145 | 605.70 | p |

a - ref. 46

b - ref. 47

c - ref. 24

p - present

TABLE 3. COMPARISON OF σ_0 VALUES AT 6943ÅUNITS: 10^{-28} cm^2

| Molecule | Extrapolated From Measured Value At | | Measured Value at 6943Å | | | EXTRAPOLATED FROM THEORETICAL VALUE DETERMINED FROM INDEX OF REFRACTION GIVEN IN TABLE 2 |
|-----------------|-------------------------------------|---------------------------|-------------------------|----------------------|-----------------------|--|
| | 3638Å Present | 6328Å Trapy et al ref. 26 | Rudder et al ref. 22 | George et al ref. 19 | Geindre et al ref. 27 | |
| O ₂ | 1.80 | ----- | ----- | 0.9 | ---- | 1.73 |
| N ₂ | 2.14 | 2.17 | 2.12 | 1.04 | 2.38 | 2.07 |
| Ar | 2.07 | 1.87 | 1.88 | 0.843 | 2.13 | 1.85 |
| Ne | 0.091 | 0.106 | ----- | 0.048 | 0.106 | 0.106 |
| He | 0.032 | 0.03 | 0.296 | ----- | 0.033 | 0.03 |
| H ₂ | 0.431 | 0.440 | 0.438 | ----- | ----- | 0.41 |
| CO ₂ | 6.35 | 5.09 | ----- | 2.76 | ----- | 4.84 |
| CH ₄ | 4.59 | ----- | 4.56 | ----- | ----- | 4.63 |

TABLE 4. RELATIVE RAYLEIGH SCATTERING CROSS SECTION

| Wavelength (Å) | ref. 22 | From Measured Values Present | | | | | ref. 31,33 | ref. 35 | Calculated from Table 2 |
|-----------------|---------|------------------------------|--------|--------|--------|--------|------------|---------|-------------------------|
| | 6943 | 6328 | 5145 | 4880 | 4579 | 3638 | 1215.7 | 1215.7 | 5893 |
| Molecule | | | | | | | | | |
| O ₂ | | 57.0 | 56.8 | 56.6 | 56.7 | 56.7 | ----- | ----- | 56.5 |
| N ₂ | 71.6 | 62.0 | 65.0 | 63.1 | 70.2 | 67.4 | 200.0 | 153.3 | 67.6 |
| Ar | 63.5 | 57.5 | 63.3 | 62.9 | 68.6 | 65.1 | 182.0 | 156.2 | 60.5 |
| Ne | -- | 2.86 | 2.86 | 2.86 | 2.83 | 2.86 | 4.3 | ----- | 3.5 |
| He | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| H ₂ | 14.8 | 13.64 | 13.5 | 13.6 | 13.6 | 13.6 | 62.9 | 65.1 | 13.4 |
| CO ₂ | -- | 201.4 | 200.0 | 200.0 | 200.2 | 200.2 | ----- | ----- | 158.1 |
| CH ₄ | 154.1 | 145.5 | 144.3 | 144.3 | 144.7 | 144.7 | ----- | ----- | 151.2 |
| Freon-12 | | 1140.0 | 1132.4 | 1132.1 | 1135.0 | 1135.3 | ----- | ----- | |
| Freon-13B1 | | 781.8 | 776.2 | 776.4 | 778.3 | 782.8 | ----- | ----- | |
| Freon-14 | | 155.5 | 154.3 | 154.3 | 154.4 | 154.7 | ----- | ----- | |
| Freon-22 | | 693.6 | 688.6 | 688.6 | 690.3 | 690.3 | ----- | ----- | |

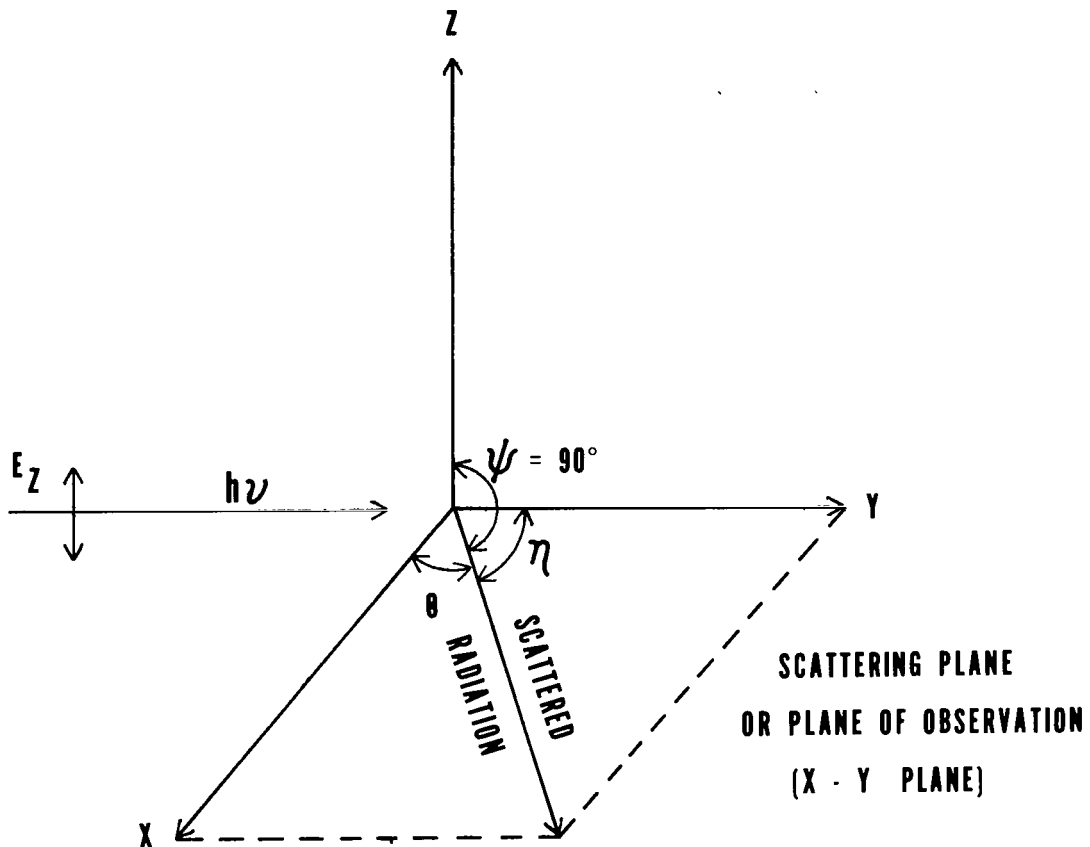


Figure 1. Geometry of light scattering: The radiation is scattered in the direction θ , η , ψ . The incident radiation is along the Y-axis. The X - Y plane is the plane of observation and $\psi = 90^\circ = \theta + \eta$.

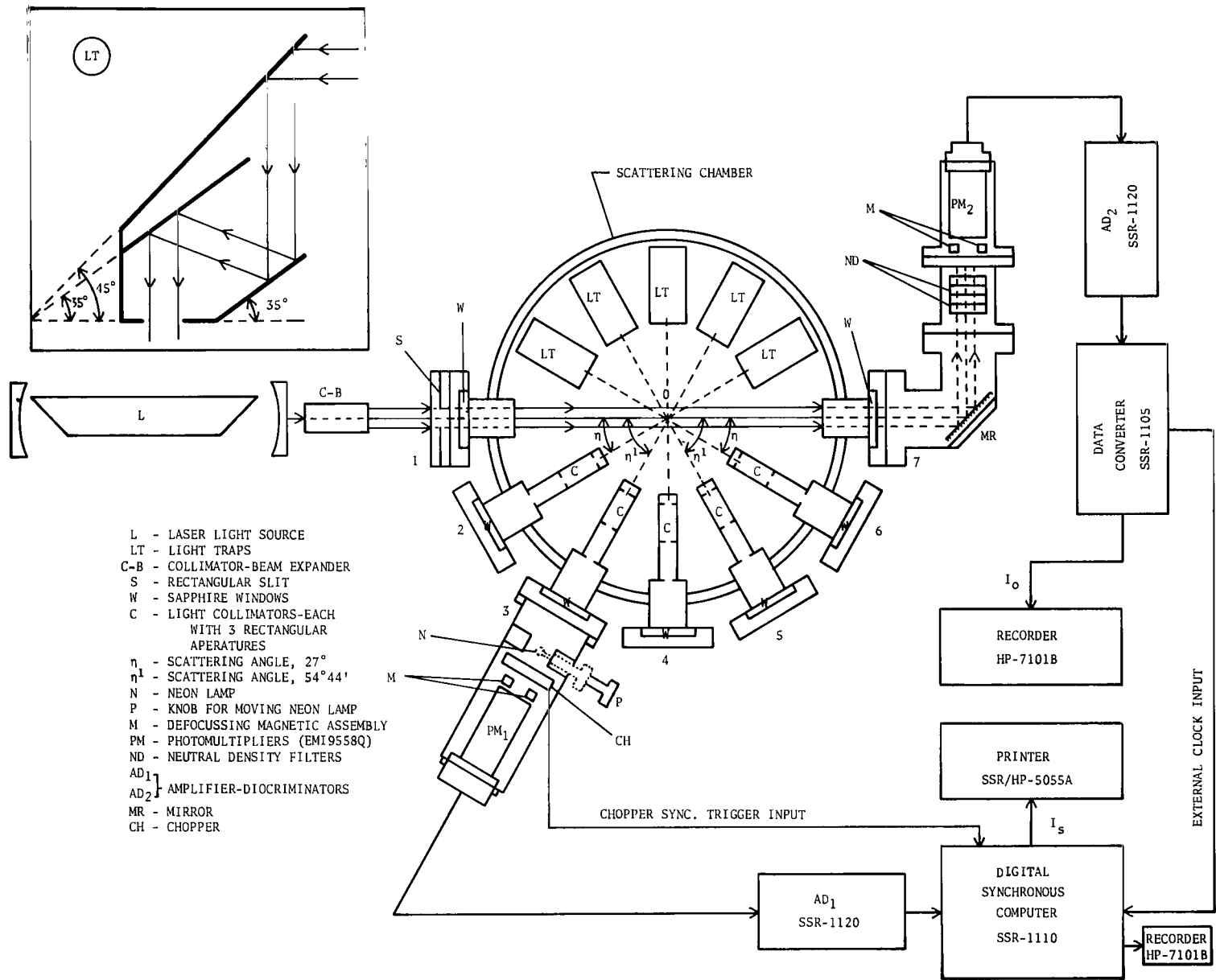


Figure 2. The block diagram of the present experimental setup.

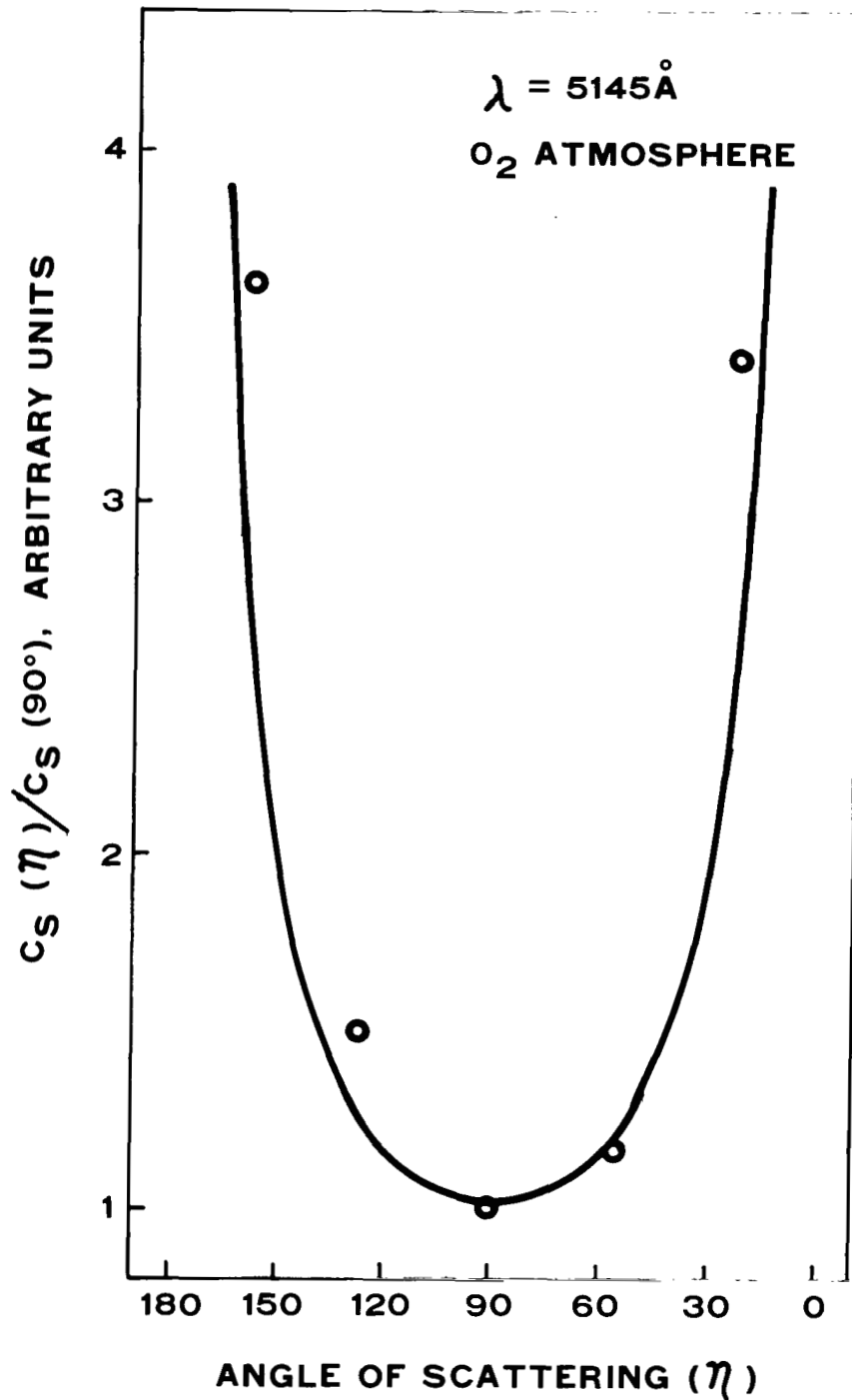


Figure 3. Intensity response of the photomultiplier as a function of angle in O_2 atmosphere. Solid curve represents the symmetry expected from molecular scattering whereas the circle points are the measured data points.

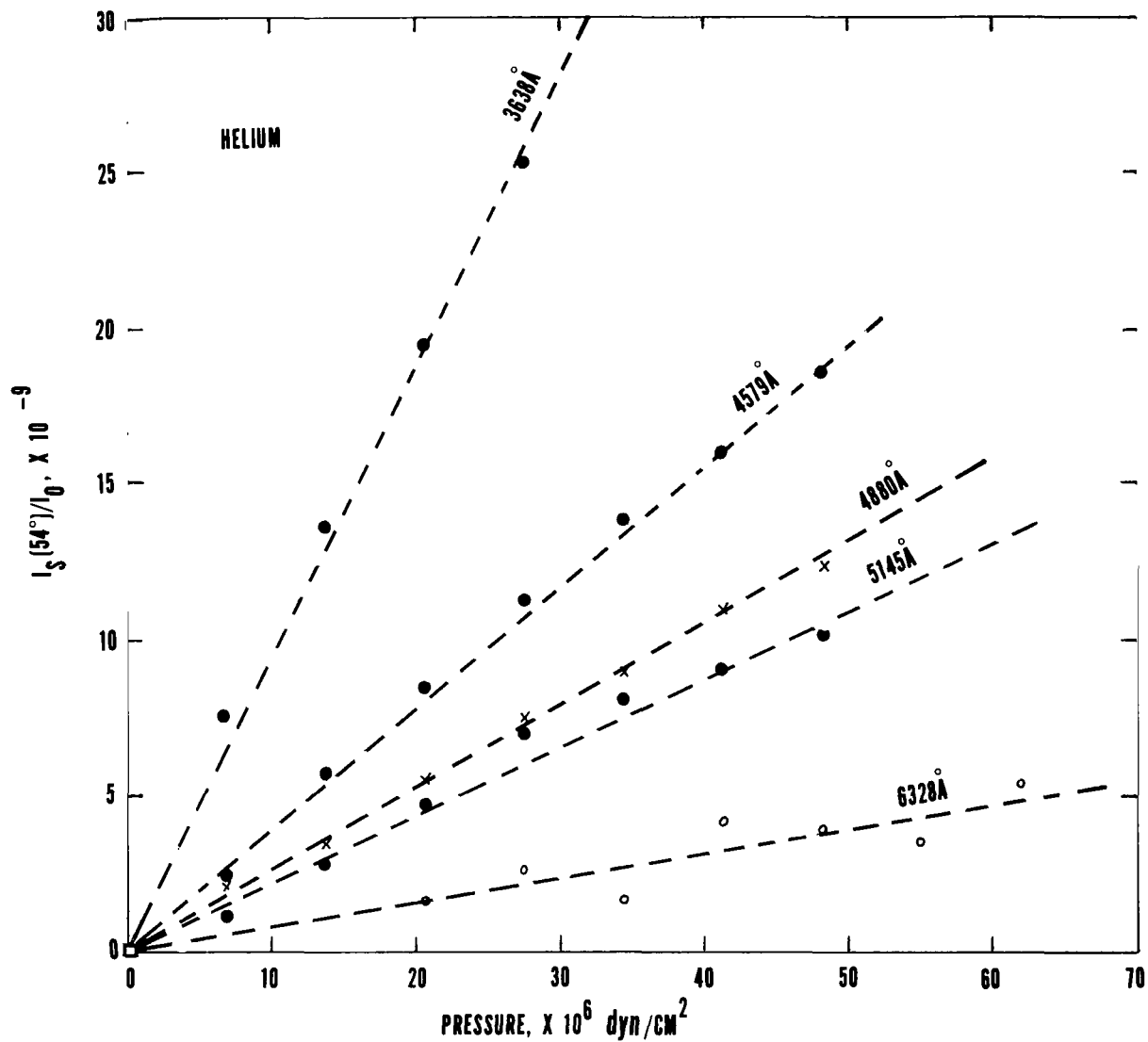


Figure 4. The ratio of 54° 44' scattering radiation to incident radiation corresponding to different wavelengths for helium as a function of gas pressure. Data Point marked \square is common to all lines.

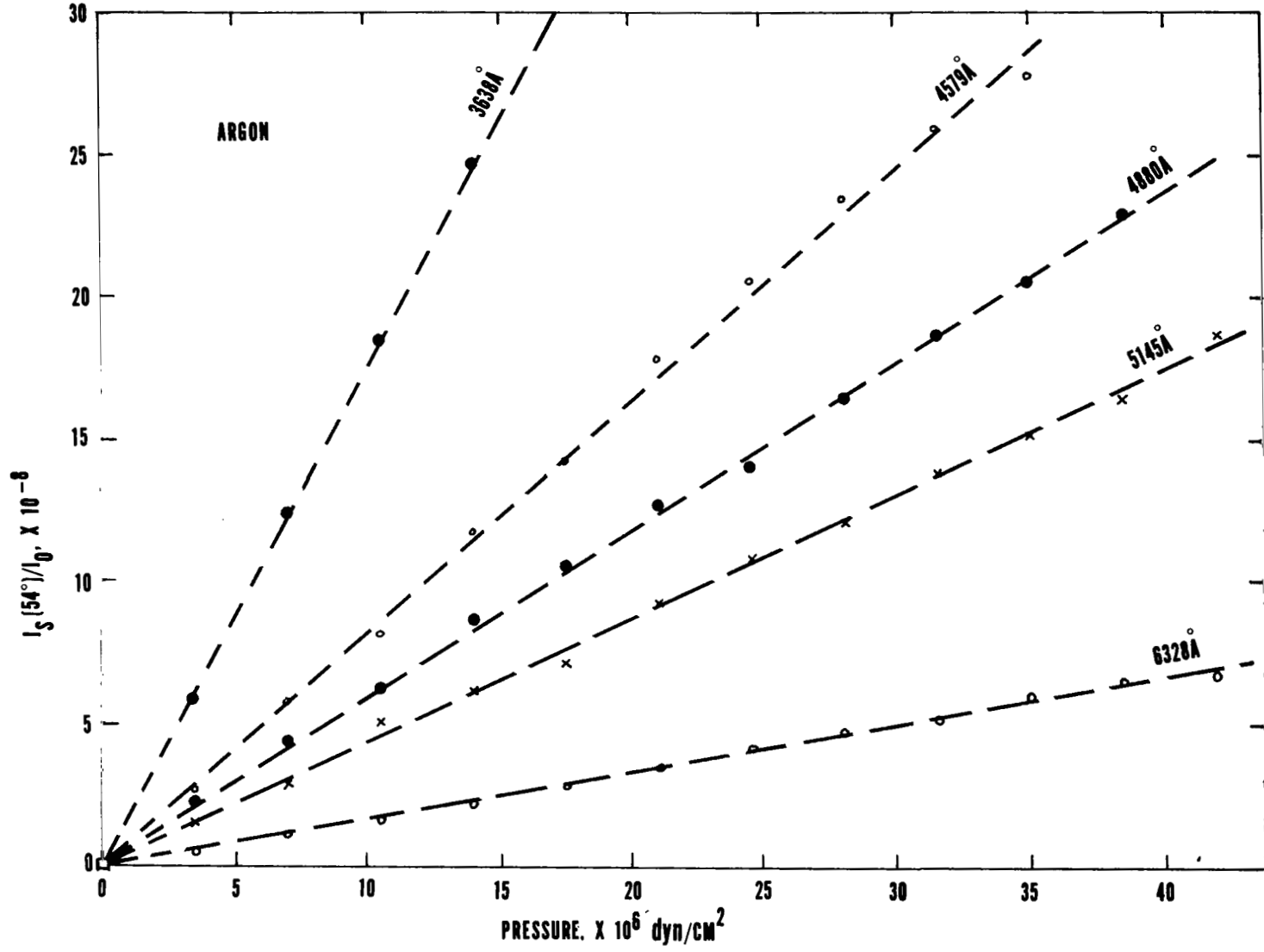


Figure 5. The ratio of $54^\circ 44'$ scattering radiation to incident radiation corresponding to different wavelengths for argon as a function of gas pressure. Data point marked \square is common to all lines.

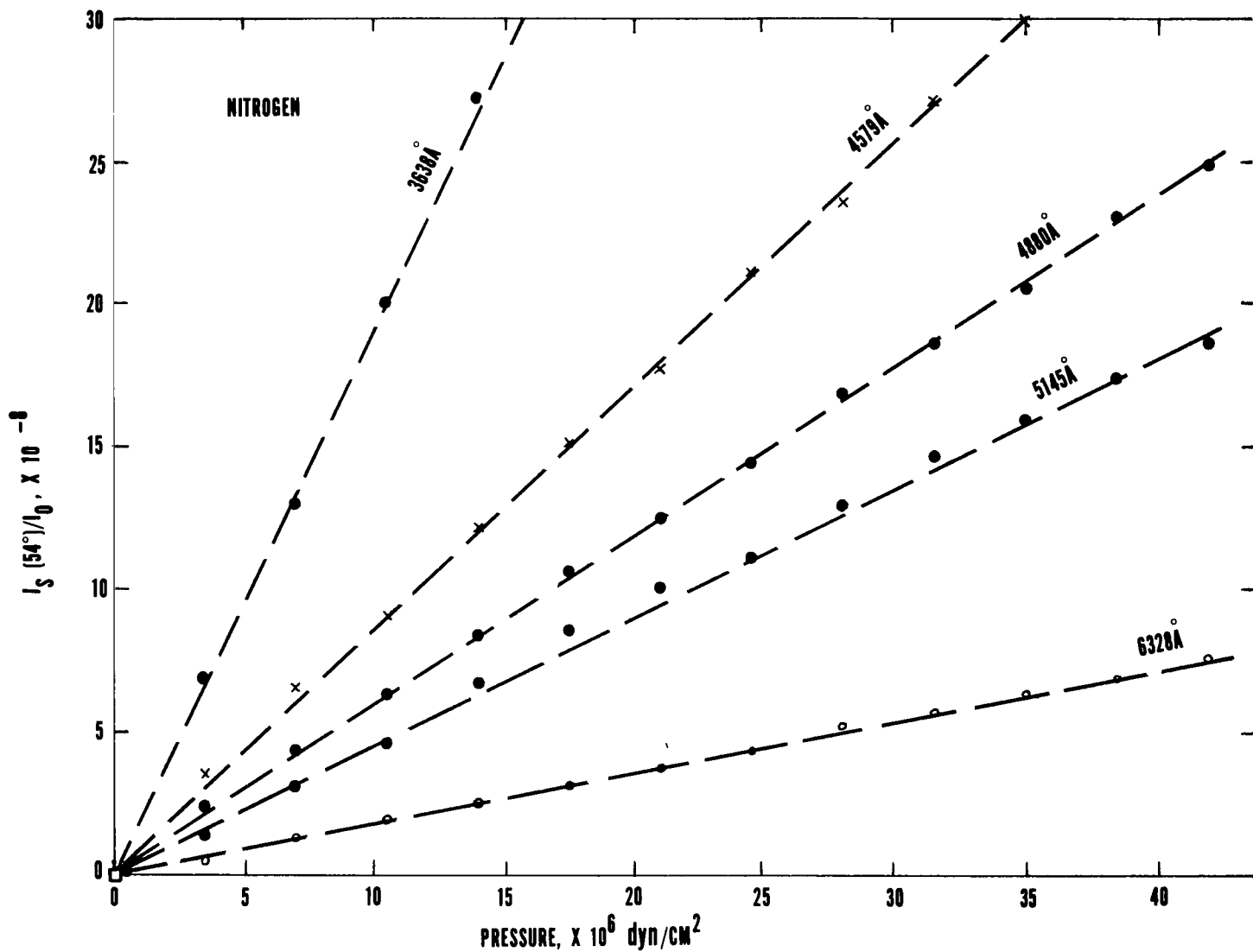


Figure 6. The ratio of $54^\circ 44'$ scattering radiation to incident radiation corresponding to different wavelengths for nitrogen as a function of gas pressure. Data point marked \square is common to all lines.

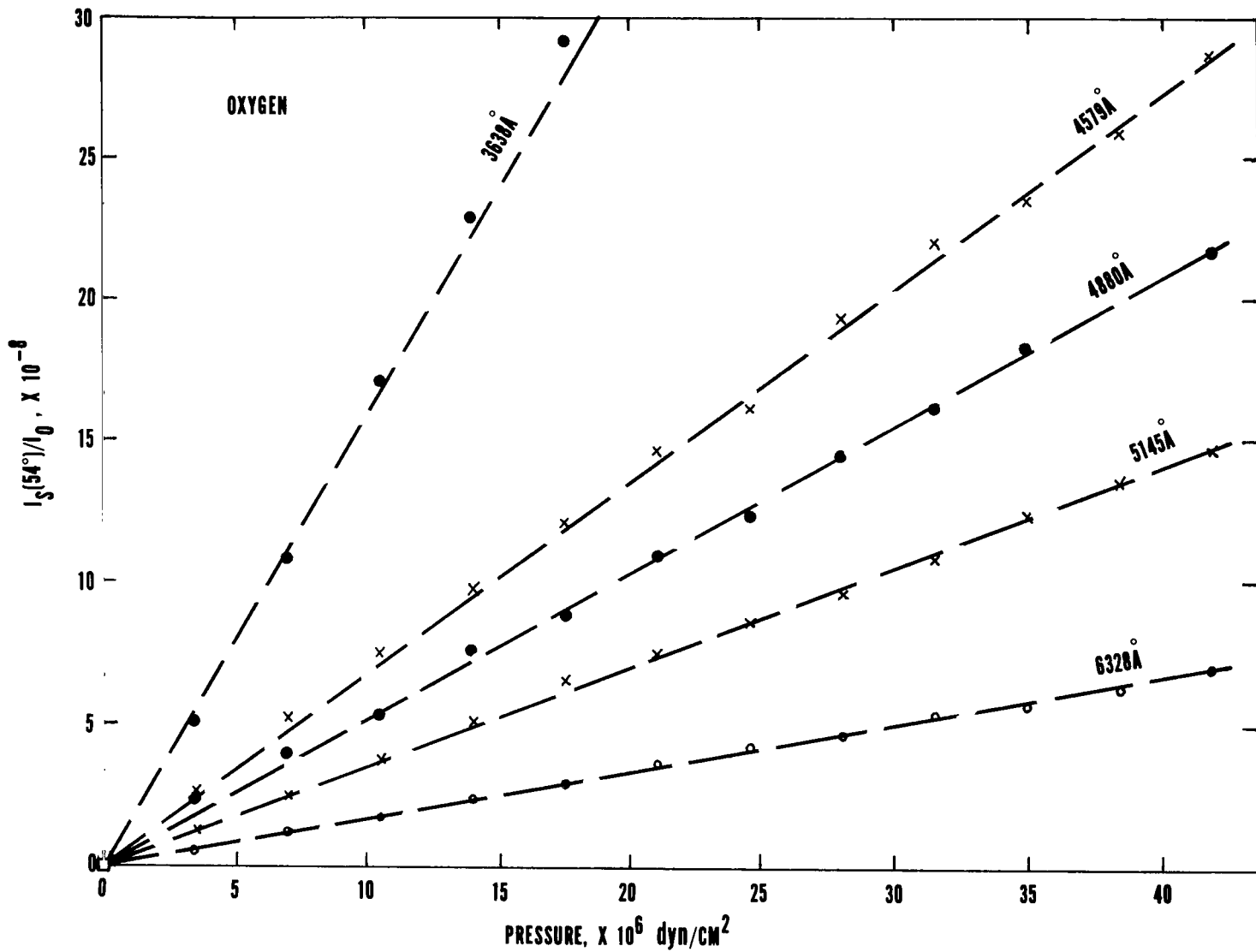


Figure 7. The ratio of $54^\circ 44'$ scattering radiation to incident radiation corresponding to different wavelengths for oxygen as a function of gas pressure. Data point marked \square is common to all lines.

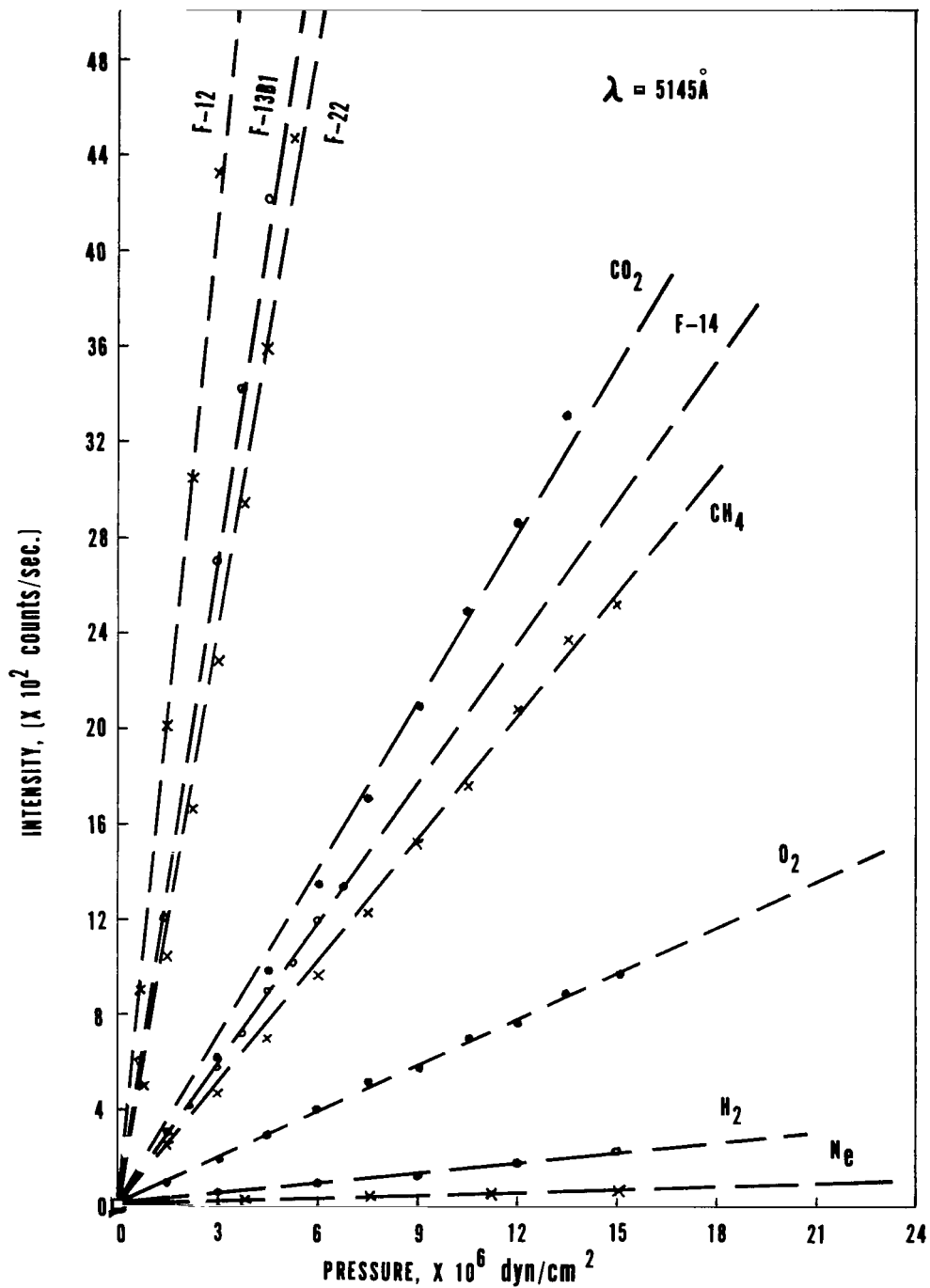


Figure 8. Photomultiplier response from $54^\circ 44'$ scattering of 5145\AA radiation for Ne, H_2 , O_2 , CO_2 , CH_4 , freon-12, freon-13B1, freon-22, and freon-14 as a function of pressure. Note that O_2 measurements are for relative comparison. Data point marked \square is common to all lines.

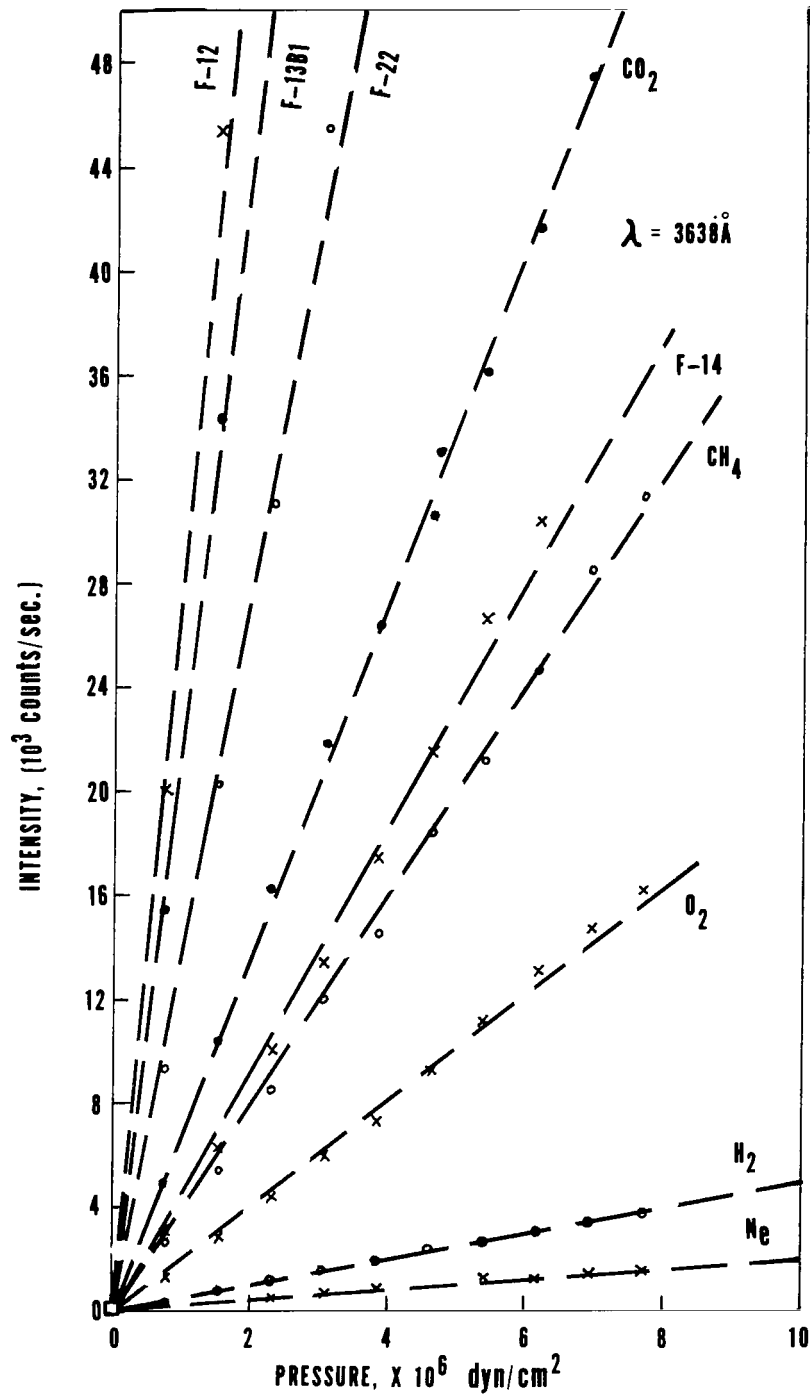


Figure 9. Photomultiplier response from $54^\circ 44'$ scattering 3638\AA radiation for Ne, H₂, O₂, CO₂, CH₄, freon-12, freon-13B1, freon-22, and freon-14 as a function of pressure. Note that the O₂ measurements are for relative comparison. Data point marked \square is common to all lines.

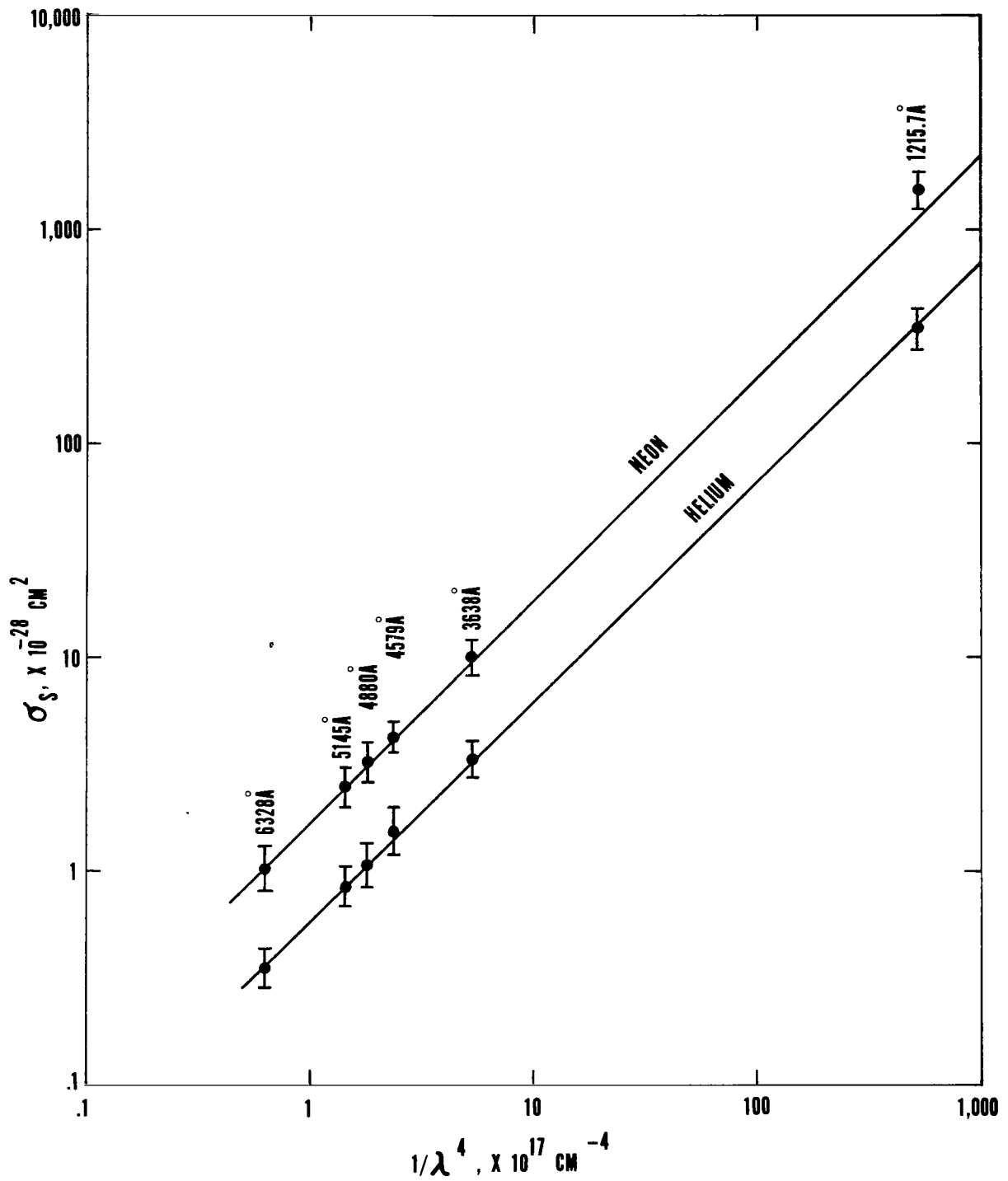


Figure 10. Variation of the measured values of Rayleigh scattering cross section with $1/\lambda^4$ for helium and neon. Note that the value of Ne corresponding to 1215.7Å does not fall on the straight line.

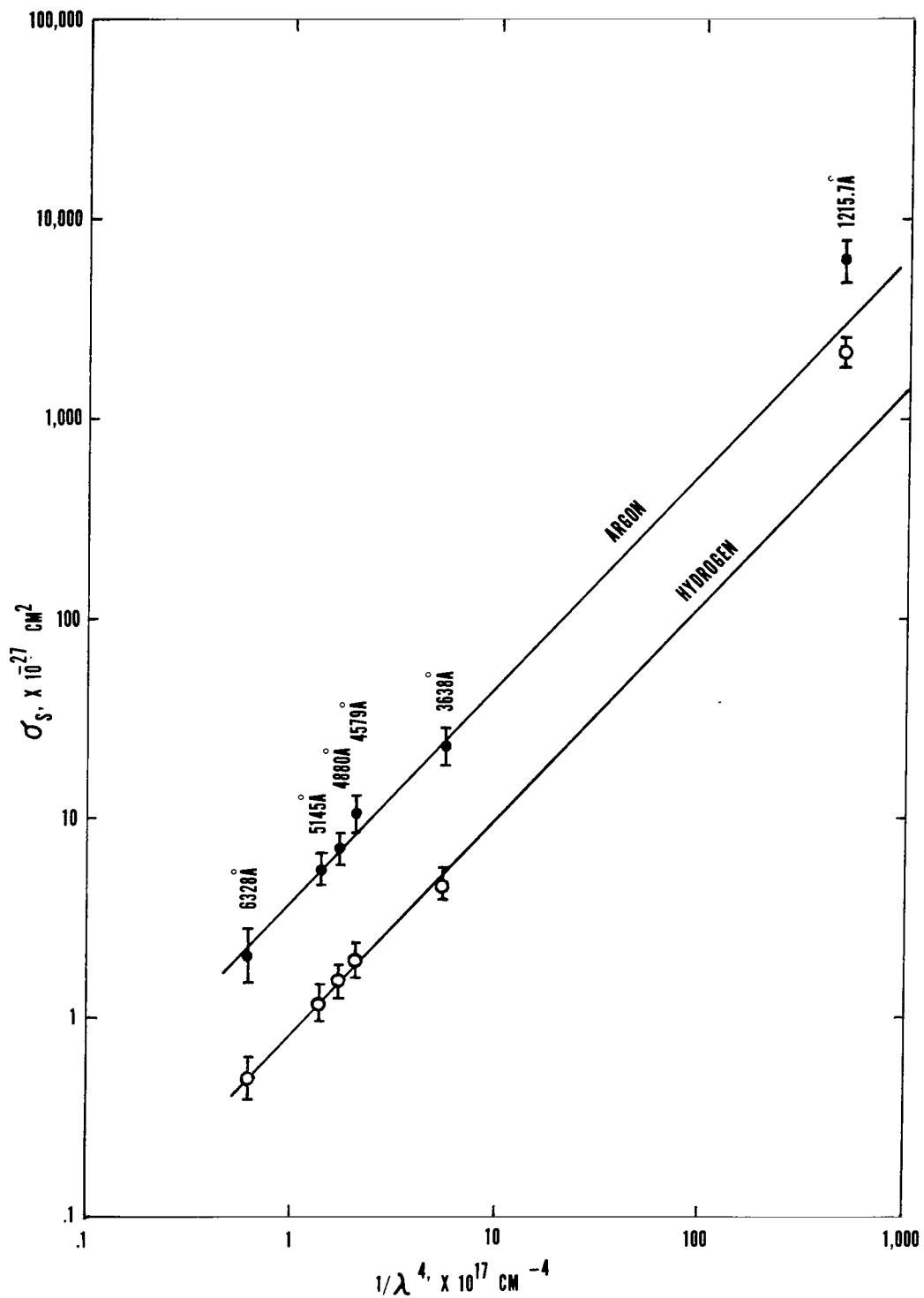


Figure 11. Variation of the measured values of Rayleigh scattering cross section with $1/\lambda^4$ for argon and hydrogen. Note that the values corresponding to 1215.7Å do not fall on the straight lines.

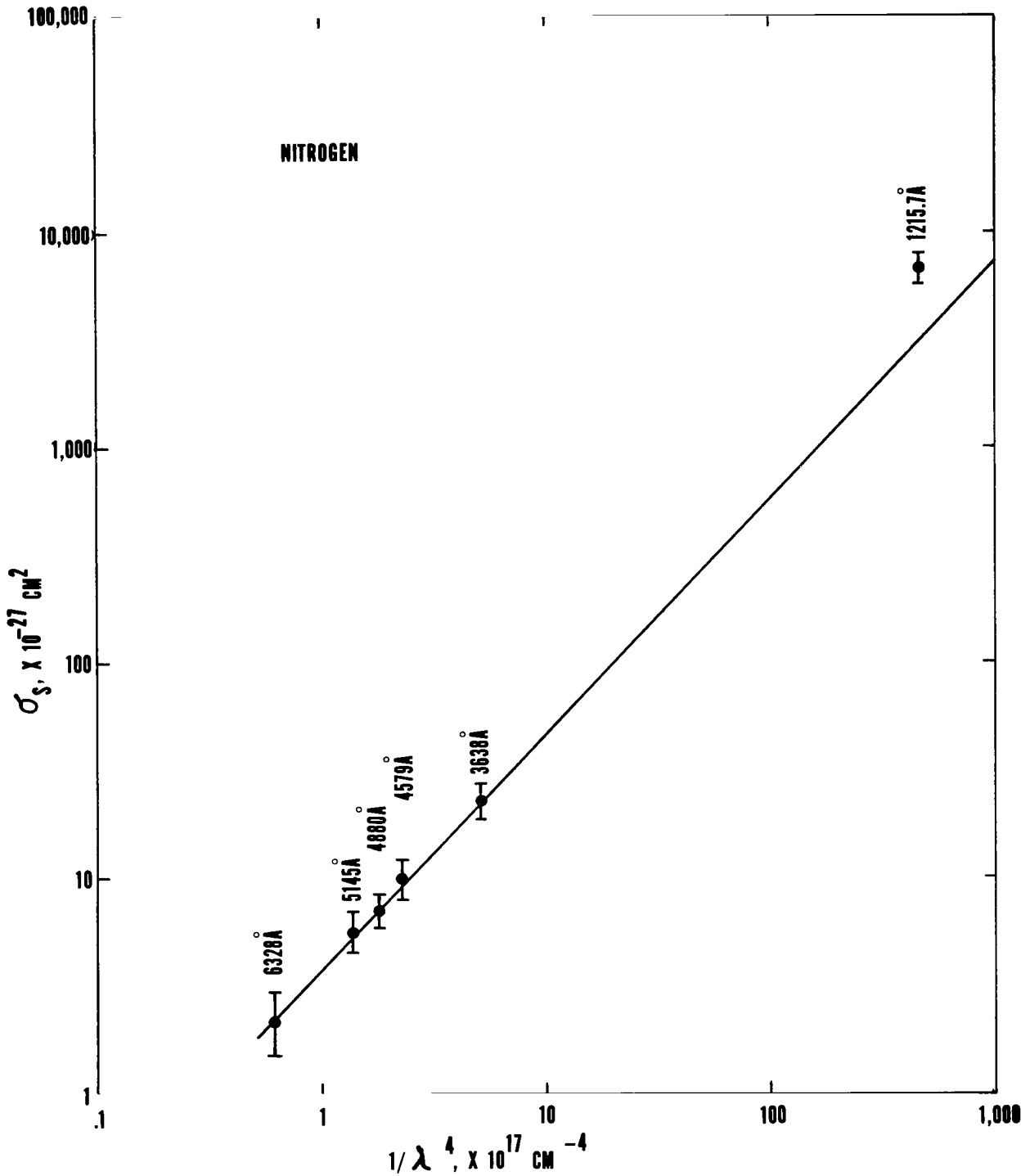


Figure 12. Variation of the measured values of Rayleigh scattering cross section with $1/\lambda^4$ for nitrogen. Note that the value corresponding to 1215.7 Å does not fall on the straight line.

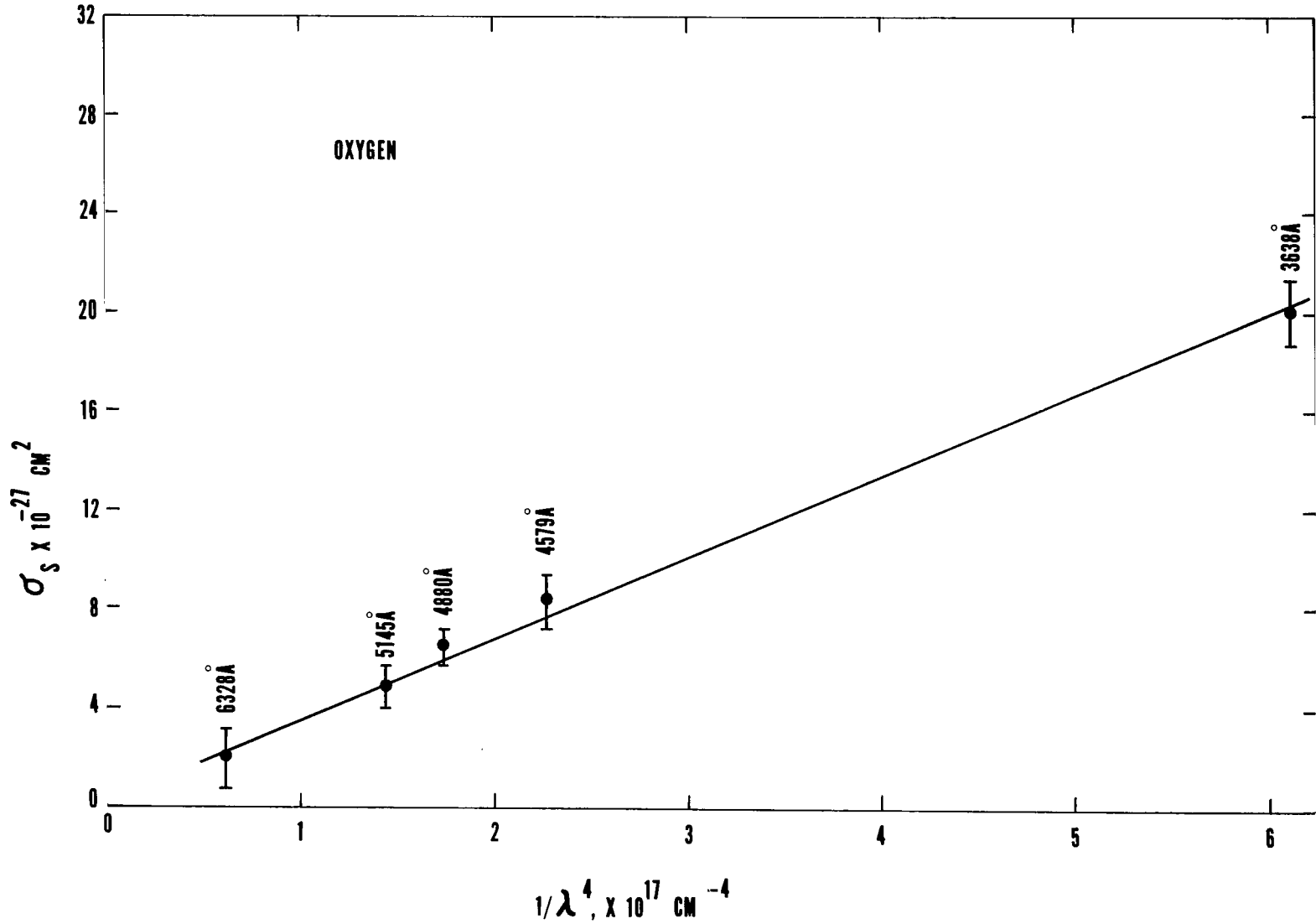


Figure 13. Variation of the measured values of Rayleigh scattering cross section with $1/\lambda^4$ for oxygen. Note that the value corresponding to 1215.7\AA is not known and therefore, not shown.

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APPENDIX I

ERROR ESTIMATES IN THE MEASUREMENTS

The parameters and their measured accuracy which may influence the total accuracy of the Rayleigh scattering cross sections are tabulated as follows:

| Parameters | Standard Deviation % Error and Remarks |
|--|--|
| C _s | ± 9 |
| C _o | ± 2.5 |
| N | ± 1.0 |
| V | ± 5 |
| Multiple reflections arising from neutral density filter | The intensity after two successive reflections was calculated to be about 0.01% of the incident intensity. Therefore, its contribution is negligible. |
| Transmission of light through sapphire windows | The transmission of light varied from one window to the other by less than 0.1% and was assumed to be the same for all windows. |
| Absorption by molecules and/or impurities | The contribution due to these are accounted for as the pathlengths from the scattering volume to the windows are the same in the experiment. |
| Background signal | The background signal was measured in vacuum conditions (pressure < 10 ⁻⁴ mm-Hg referred as zero pressure in the text) and then subtracted from each observation. The background signal was measured before and after each set of measurements. The variation was negligible. |

Examination of the table shows that the error in the values of σ_s will depend on the accuracy of the values of C_s , C_0 , N and V . From the equation (5) it may be shown that

$$\sigma_s = 4\pi \frac{I_s}{I_0 NV} .$$

Since $I_s/I_0 = C_s/C_0$, the above expression can be written as

$$\sigma_s = 4\pi \frac{C_s}{C_0 NV} .$$

Therefore, the error in the values of σ_s can be calculated from the expression

$$\left[\frac{\sigma(\sigma_s)}{\sigma_s} \right]^2 = \left[\frac{\sigma(C_s)}{C_s} \right]^2 + \left[\frac{\sigma(C_0)}{C_0} \right]^2 + \left[\frac{\sigma(N)}{N} \right]^2 + \left[\frac{\sigma(V)}{V} \right]^2$$

Using the error values shown in the table, the value of the right hand side is found to be 0.011325. This gives the error of $\leq 11.0\%$ in the values of σ_s reported in this study.

Note - σ_s scattering cross section
 σ standard deviation

APPENDIX II

ERROR ESTIMATE DUE TO THE PRESENCE OF PARTICULATE MATTER

According to Van de Hulst (ref. 48) the ratio of forward to backward scattering may be expressed as

$$\frac{I_s(\eta = 0)}{I_s(\eta = 180)} = 1 + \frac{4X^2}{15} \left[\frac{(\mu^2 + 4)(\mu^2 + 2)}{(2\mu^2 + 3)} \right]$$

This relation holds good for $X < 1.0$. For Rayleigh scattering the ratio $I_s(\eta = 0)/I_s(\eta = 180)$ is unity. In the present experiment the particulate matter of the size greater than $25 \text{ m}\mu$ is filtered. Therefore, particles of size smaller than $25 \text{ m}\mu$ are still present. Let us calculate asymmetry that might be caused by the particles of the size $25 \text{ m}\mu$. This has been calculated for a wavelength 5145\AA . Assuming the index of refraction of these particles as $\mu = 1.55$, the value of $X = \pi D/\lambda$ would be 0.1526 . Here D is the diameter of the particles which is assumed to be $25 \text{ m}\mu$. Using these quantities, we find

$$\frac{I_s(\eta = 0)}{I_s(\eta = 180)} = 1.022$$

This means that an enhanced asymmetry in the forward direction would be 2.2% .



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