THE SCATTERING OF OPTICAL RADIATION FROM AIRBORNE AEROSOLS

A Dissertation

Presented to

the Faculty of the Department of Electrical Engineering

University of Houston

In Partial Fulfillment

of the Requirements for the Degree

Doctor of Philosophy in Electrical Engineering

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by

Jagdishchandra T. Gajjar

August, 1970

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ABSTRACT

In the study of optical radiation scattered from aerosols a controversy has existed as to whether the scattered radiation that results when the incident radiation comes from a laser is different from the radiation which results when the source of incident radiation is a linearly polarized, narrowband thermal source. An experimental system is developed to investigate the nature of the scattered radiation which employs photo-electron counting. It is argued that the scattered radiation may be neither coherent nor totally incoherent. A model for the scattered radiation is constructed from physical considerations which consists of a sum of coherent and incoherent components. Normalized second and third factorial moments of the counting distributions are computed from the experimental data which is obtained by using scattered laser light, scattered thermal light and radiation from a light emitting diode as the radiations illuminating the photomultiplier tube. These are compared and their behaviour with respect to the average intensity is analysed and is explained, and the applicability of the model is discussed. It is concluded that the radiation reaching the detector does contain a small coherent component if the radiation incident upon the

scatterers comes from a laser. A few examples are presented showing the significance of the findings on the interpretation of experimental results in light scattering experiments.

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LIST OF SYMBOLS

A	Modulating signal
F(k)	k th factorial moment of the photo-counting distribution
I	Intensity at detector surface (scaler component)
I_c	Intensity of coherent component
I_i	Intensity of incoherent component
I_o	Ensemble average of $<I_i>$
M	Total number of counts
N	Number of scatterers in scattering volume
N(k)	Unbiased estimate of F(k)
T	Counting period
U	Integrated intensity
U_c	Coherent integrated intensity
U_i	Incoherent integrated intensity
U_o	Ensemble average of U_i
V	Component of the electric field at the detector in the plane of polarization determined by the linear polarizer placed in front of the detector
V_c	Amplitude of the "coherent component" of V
V_i	Amplitude of the "incoherent component" of V
V(k)	Variance of N(k)
a, a', b, b', c, c'	Polynomial coefficients
a_i	Distance of the center of the i th scatterer from a plane perpendicular to the direction of progagation and passing through the center of the scattering volume

$e_o(t)$ Amplitude modulated signal

n Number of photo-electrons

n_p Number of counts recorded in the p th sampling period

$\bar{n}$ Average number of counts

$\vec{r}$ Position of detector relative to the scattering volume

t Time

w_c Carrier frequency

α Detector sensitivity
Ratio σ_u/U_o

α_i Phase of the radiation reaching the detectors from the i th scatterer relative to the phase of the incident beam

β Ratio of average value of U_c to U_o

θ Angle between direction of propagation and the lines joining the detector to the scatterers

ϕ Angle between direction of polarization of the incident beam and the lines joining the detector to the scatterers

Φ Amplitude of the electric field scattered from each scatterer, at the detector

ψ Electric field at the detector which is the sum of the fields scattered coherently from all the scatters

ω Radian frequency of the incident radiation

σ_i Standard deviation of the distribution of $\langle I_i \rangle$

$\sigma_N(k)$ Standard deviation of the distribution of the normalized k th factorial moments

σ_u Standard deviation of the distribution of U_i

τ_c Correlation period of the radiation

$\langle \ \rangle$ Ensemble average

CHAPTER I

INTRODUCTION

The scattering of light by particles and molecules has been the subject of a large number of investigations in the various scientific disciplines. This is due to the fact that light and other electromagnetic radiation interact non-destructively with most matter and hence scattering offers a fast and convenient technique of obtaining information about the object doing the scattering. The theoretical investigations that have been carried out have been based on the Rayleigh-Gans and the Mie theories. Experimental investigations performed using band limited thermal sources in the optical region and highly monochromatic sources in the microwave region have verified these theories within the limitations of the experimental procedures used.

The development of the laser has resulted in a highly monochromatic and intense source of light radiation. This development has led to renewed interest in the phenomenon of light scattering. The major advantage to be gained from the use of a laser source in experiments on light scatter lies in the high intensity and the spatial and temporal coherence of the radiation.

Theoretical work connected with the scattering of laser light has generally been an extension of the theories developed for use in radar scattering studies. Radar signals can be considered as almost pure monochromatic electromagnetic radiation in the radio and micro-wave region of the frequency spectrum. This radiation has a high degree of both spatial and temporal coherence. One important result of these radar scatter studies is the principle of the so called "incoherent scattering". According to this concept, the average intensity at a detector due to the sum of the radiation scattered from randomly placed scattering particles will be the same whether the illuminating source is coherent or non-coherent.

This theory is based upon the supposition that the square of the amplitude of the sum at the signals from randomly placed scatterers will be the same as the addition of the intensities of the illumination of the detection aperture. This assumption is based upon the lack of definite phase relationships among the individual waves scattered from the different particles.

An experiment upon the measurement of the scattering functions from artificially generated fogs using both the He-Ne laser and a tungsten lamp source was reported by Carrier and Nugent[1]. The scattering functions obtained from the two sources differed by greater amounts than can be conveniently explained from theoretical considerations. This experiment can either be interpreted as casting doubt upon the theory of incoherent scattering, or maybe the speculation arises that there is a basic difference in the manner of light scattered from a laser light source and that observed from a thermal light source.

Other investigators who have made measurements of this same nature include Harris, Sherman and Morse[2], and Reisman, Cumming and Bartky[3]. These measurements show no significant differences between scattering from laser sources and thermal sources for the Mie region. The experiment by Harris et. al. used mono-disperse hydrosols for scatterers. The experiment by Reismann et. al. used artificially generated fogs. The results of both investigations showed complete agreement with the Mie theory.

More recently an experiment has been reported by Zuev, Kabanov and Savelov[4]. This experiment compares the attenuation coefficients of laser light and thermal light when both were transmitted through artificial fogs and smokes. No appreciable differences between the attenuations was observed. A small difference between the measured values and those predicted by the Bouger-Beers law was considered as being due to multiple scattering effects. An additional experiment by Setzer[5] has been reported. In this experiment a comparison between the scattering functions for particles in an aerosol with that predicted from the Mie theory shows close agreement also.

In the Rayleigh scatter region an experiment by Georgi, Yokohama, Goldstein and Slama[6] has been published. These investigators have compared the angular dependence of laser light scattered from Xenon atmosphere with the values calculated using Rayleigh theory. A considerable difference between the measured and calculated values was found to exist. Later measurements made by Watson and Clarke[7] and by Leite, Moore, Porto and Ripper[8] shows complete agreement with the Rayleigh theory.

If the predominant theme of the previously mentioned articles is examined it becomes obvious that some fundamental questions concerning the scattering from laser light sources have not been fully answered. In general one might conclude that the Mie theory and the Rayleigh theory are probably valid for the prediction of scattering for the laser light sources. In most of the cases reported to date little difference seems to exist between the use of the laser light source and a monochromatic narrowband gaussian light source such as a thermal source as the source of incident radiation. All of the experiments mentioned have one common factor. In each case the detection measurement has been the photo-multiplier tube anode current. This current is in reality a measurement proportional to the time average rate of electron arrival at the anode. In order to minimize the effects of large fluctuations in the instantaneous value of this current, it becomes necessary that a large period of integration be used. The period of the integration and the uncertainty in the value of the current being measured are inversely proportional. This pre-supposes the signal intensity on the photo-cathode surface being a constant over the period of integration. In addition the integration time and the intensity of the illumination must be selected so that a large number of cathode events takes place during the period. These constraints require that scattering occur from a population consisting of a large number of identical small particles so one may invoke the principle of incoherent scattering . This is especially true where the incident radiation is of short correlation time. The other case in which such measurements are suitable is that of a single large particle illuminated by light of high intensity.

In addition to the photo-multiplier detection problems cited, some of the experiments have used hydrosols. In these cases the background Rayleigh scattering intensity is not necessarily small compared to the scattering intensities at the cathode surface resulting from the Mie scatterers. Reflections and scattering from chamber walls have also been neglected when computing experimental errors.

If we wish to consider the scattering from a population of aerosols, where multiple scattering effects may be neglected, it becomes evident that the number of individual scatterers in the illuminated volume will be a time variant quantity. As the particles move through the volume, time variant interference effects will create a random fluctuation in the signal intensity at the detector surface. The signal at the surface of the detector cathode is thus a random time function. In order to compare the scatter from aerosols by laser light to that from thermal light, the total statistics of such a process must be considered. The mean value alone, which is the value measured in an integrated current measurement, is not sufficient to show any differences which might exist.

A comparison of the probability distribution of the number of counts occuring during a period T when a detector is illuminated by an amplitude stabilized laser to the distribution obtained during T for illumination by a gaussian thermal source has been made. By using Mandel's formula[9], Sudarshan and Klauder[10] have shown that if the correlation time of the laser is long compared to the counting period, a Poisson distribution will be obtained for the laser source, while the polarized narrow band thermal source with a short correlation time will yield a Bose-Einstein distribution. The above has been experimentally verified by Arecchi[11].

It is the purpose of this investigation to attempt to achieve experimental evidence to answer the following questions:

(a). How does scattering modify the counting distributions and the distribution of the field intensity on the surface of the detector as a function of the angular position of the detector relative to the direction of the incident radiation?

(b). Can any more information be obtained about the system doing the scattering from a consideration of the higher order counting moments?

(c). Is there any advantage to using the laser as a source in scattering experiments?

In order to obtain this evidence we perform an experiment wherein the aerosol is made to flow in a free jet and is contained within a sheath of clean dry air. Using a detector that is mounted so that it can be moved concentric with the aerosol column, pulse counting distributions are obtained for scatter from both laser and thermal radiation. These are then inverted to obtain the distributions of the field fluctuations at the detector surface. The next chapter presents some theoretical complements. This is followed in chapter three by a description of the experimental apparatus. The experimental procedure and computational techniques are elaborated upon in the next chapter. The last chapter presents and discusses the results.

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CHAPTER II

THEORETICAL COMPLEMENTS

This chapter presents the theoretical background upon which the present investigation is based. Although much of the material presented here is not new, it seems desirable to present it in a consolidated form. The first part of the chapter deals with the detection process. For the present purpose a semi-classical treatment is presented since it is simpler and physically more meaningful than the exact quantum mechanical treatment. A few counting distributions are next arrived at from theoretical considerations. The last part of this chapter concerns some computational aspects of this investigation.

Consider a uniformly illuminated detector illuminated with a time varying intensity of radiation I(t). It can be shown[12-15] that the probability p(n,T,t) of counting n photo-electrons in the interval t to t+T is given by the ensemble average over the Poisson distributions;

$$p(n,T,t) = \frac{1}{n!} \langle [\alpha U(T,t)]^n \exp[-\alpha U(T,t)] \rangle \qquad (1)$$

where $$U(t,T) = \int_t^{t+T} I(t')dt' \qquad (2)$$

the average being taken with respect to the ensemble of

U(t,T), and α is a parameter which is a measure of the detector sensitivity. The above may be rewritten in the more familiar form of Mandel's well known formulae:

$$p(n,T,t) = \int_0^\infty \frac{(\alpha U)^n}{n!} e^{-\alpha U} p(U)\,dU \qquad (3)$$

with α and U as defined above. Thus the information regarding the field fluctuations is contained in the statistics of U and hence in those of n. If the field is stationary as is often the case the dependence of p(n,T,t) on t may be dropped.

Two special cases may be considered. The first when I(t) does not fluctuate significantly in time. This situation is approximated by a stabilized single mode laser beam. It can be shown in this case[16,17] that the resultant counting distribution is a Poisson distribution. The other case which readily lends itself to analysis is that in which the field is a Gaussian random process and the measurement time T is short compared to the correlation time of the light. In this case the resulting counting distribution can be expressed as[16,17];

$$p(n,T) = 1/\{(1+\langle n\rangle)[1+(1/\langle n\rangle)]^n\} \qquad (4)$$

which is the familiar Bose-Einstein distribution.

In a multiparticle scattering process the scattered radiations from each individual particle interact to give the resultant radiation at the detector surface. The intensity at the detector is thus a random process which depends upon the incident radiation and upon the locations of the individual particles, all of which in themselves are random processes. In order to visualize the manner in which the scattering modifies the incident radiation, consider N identical scatterers, identically oriented, and situated at a large distance from the detector, such that the detector surface lies within a coherence area [18] of emission from each scatterer assumed as a point source. Consider a linearly polarized plane wave of coherent monochromatic radiation incident on these scatterers. The amplitude of the scattered radiation emitted from each scatterer is a function of the co-ordinates of the detector relative to the scattering center and of the size and location of each scatterer. Since the detector is located at a large distance from the scattering volume and the scattering volume is small it may be assumed that the angles between the direction of propagation and the lines joining a point on the detector to the individual scatterers are all nearly equal, say θ. Also the angles between the above mentioned lines and the direction of the polarization vector of the incident radiation are all nearly equal, say ϕ. Further the amplitudes of the radiation scattered from each scatterer and reaching the detector are also equal to say $\Phi(\vec{r})$; where $\vec{r}$ denotes the distance of the detector center from the center of the scattering volume. If the scatterers are assumed stationary, i.e. for times short compared to the smallest periods of the doppler shifts introduced due to the movement of the scatterers the total field at the detector is given by:

$$\psi(\vec{r},t) = \sum_{\text{all scatterers}} \Phi(\vec{r})e^{j(\omega t+\alpha_i)} \tag{5}$$

where ω is the radian frequency of the incident radiation, and α_i is the relative phase of the radiation scattered from the ith scatterer. The phase α_i depends upon $\vec{r}$, the position of the detector relative to the ith scatterer and upon a_i the distance of the ith scatterer, from a plane perpendicular to the direction of propagation which passes through the center of the scattering volume.

As a first approximation, assume that the α_i's are uniformly distributed random variables. In this case the problem of finding the possible values of $|\psi|^2$ is readily identified with the problem of a random walk in two dimensions with steps of constant length. If the number of scatterers N is large, then it can be shown[19] that the distribution of the amplitude $|\psi|$ is a Rayleigh distribution:

$$p(|\psi|) = \frac{2|\psi|}{N\Phi^2} \exp(-|\psi|^2/N\Phi^2) \, (\psi \geq o) \tag{6}$$

The intensity of the scattered field is proportional to $|\psi|^2$ and its distribution is the exponential distribution given by:

$$p(I) = \frac{1}{<I>} e^{-\frac{1}{<I>}} \tag{7}$$

where $<I> = N\Phi^2$ represents the average intensity.

Assuming that the differential velocities of the scatterers are small, i.e. the scatterers are in a laminar flow, so that the coherence time of the scattered radiation incident on the detector is large compared with the observation period, an assumption valid for observation times of a few microseconds in the case of single scattering, the photo-count distribution can be obtained using Mandel's formula of equation (3) where now U = IT; T being the observation time, and

$$p(U) = \frac{1}{T} p(I) \tag{8}$$

i.e. $$p(U) = \frac{1}{<IT>} \exp(-U/<IT>) \tag{9}$$

where <IT> is the average value of the variable U.

The resultant photocount distribution can be shown to be given by:

$$p(n) = (1 + \bar{n})^{-1} \; (1 + 1/\bar{n})^{-n} \tag{10}$$

where p(n) is the probability of obtaining n counts and $\bar{n}$ is the average number of counts. The distribution is the well known Bose-Einstein distribution. The above calculation is based on the assumption that the field on the detector surface is a gaussian field.

Due to the motion of the scatterers in the scattering volume, the intensity on the surface is a random process. The field on the surface i.e. $V(\vec{r},t)$ is also a random process, and due to the large number of scatterers involved, so that the central limit theorem applies, the field is a normal process. Furthermore, the scatterers in the scattering volume are being generated by an aerosol generator the performance of which depends only on the air pressure in the line and on the concentration of the solution from which the aerosol is obtained by atomization. Thus the number of scatterers in the scattering volume is a stationary random function. The mean value of the scattered field fluctuation is constant and its autocorrelation function depends upon the doppler frequency caused by the motion of the scatterers and by the autocorrelation of the random process denoting the number of scatterers in the scattering volume. In a laminar flow of scatterers, perpendicular to the direction of the light beam, the doppler contributions arise only from the transverse motion of the scatterers. This motion arises as a lateral Brownian motion with a small mean velocity. Since there are two degrees of freedom and the particles are small the mean energy can be approximated as kT, where k is the Boltzman constant and T is the absolute temperature. For a particle with a specific gravity of 1.0, having a diameter of 1 micron the mean velocity is approximately 3.9 mm/sec. In a counting interval of a few micro-seconds, the average movement of the particles will be much less than one wavelength and hence the resulting doppler effect may be neglected as long as the light reaching the detector is scattered only once from the scatterers. In the case of multiply scattered radiation, however, the situation is considerably different. In this case doppler shifts will be introduced by the inter-scatterer propagation which can have a large component along the direction of the laminar velocity flow of the scatterers. It may be noted that due to the parabolic distribution of the velocities across the cross section of the free jet, the differential velocities in this direction are not necessarily small and considerable doppler shifts may result. Indeed, if multiple scattering accounts for a considerable portion of the energy received at the detector, the simple models that have been described earlier will no longer apply and the whole process will become complicated by the introduction of a near incoherent term caused by multiple scattering. In this case a new model may be constructed as described in the following discussion.

The radiation reaching the detector may be assumed to consist of two components, the "incoherent component" which by definition has its correlation time τ_c much smaller than the counting period and the "coherent component" which is defined to have its correlation period much larger than the counting period. The photo-electron counting distribution depends upon the integrated field intensity as can be seen from equation (3). Thus if V_c represents the coherent component of the field amplitude and V_i represents the incoherent component of the amplitude on the detector surface, then the total amplitude V is given by:

$$V = V_i + V_c \qquad (10)$$

The field intensity I is given by:

$$I = V^2 = (V_i + V_c)^2 \qquad (11)$$

and the integrated field intensity U is given by:

$$U = \int_{t}^{t+T} (V_i^{\,2} + V_c^{\,2} + 2V_iV_c)\,dt' \tag{12}$$

where T denotes the counting period. The following assumptions are made.

Since the coherent and the incoherent components arise due to single and multiple scattering, respectively, they are assumed independent of each other. The coherent component of the field amplitude V_c is the sum of a relatively large number of independent contributions which are uniformly distributed phasors. The incoherent component of the intensity depends upon the number of scatterers present in the scattering volume. Finally, the average number of scatterers in the scattering volume is assumed to be large so that their number is a normally distributed, stationary, stochastic function having a correlation time long compared with the counting period T, and of the order of or less than the intersample interval.

The following definitions are next made. U_i, the incoherent integrated intensity is defined by:

$$U_i = \langle I_i \rangle\, T \tag{13}$$

where, T represents the counting period and $\langle I_i \rangle$ represents the average of the incoherent intensity $I_i = V_i^2$ over the period T. U_c, the coherent integrated intensity is given by:

$$U_c = |V_c|^2 T = I_c T \tag{14}$$

where T, as before, represents the counting period and I_c represents the coherent intensity, assumed constant over the counting interval T.

The number of particles in the scattering volume in different counting periods will vary and so will the mean incoherent intensity and the integrated incoherent intensity, since these are assumed to be directly proportional to it. The distribution of the mean incoherent intensity $<I_i>$, hence will also be normal and may be expressed as:

$$p(<I_i>) = \frac{1}{\sigma_i\sqrt{2\pi}} e^{-\frac{(<I_i>^2 - I_o)^2}{\sigma_i^2}} \tag{15}$$

where I_o is the ensemble average of the incoherent intensity and σ_i^2 represents the variance of $<I_i>$ about its mean I_o. Similarly, the distribution of the incoherent integrated intensity U_i may be represented by:

$$p(U_i) = \frac{1}{\sigma_u\sqrt{2\pi}} e^{-\frac{(U-U_o)^2}{\sigma_u^2}} \tag{16}$$

where U_o represents the ensemble average of U_i and σ_u^2 denotes the variance of U_i about its mean U_o.

At this point, in order to clarify the terminology used, it may be advantageous to employ the analogy of an amplitude modulated signal

$$e_o(t) = A(t) \operatorname{Cos} w_c t;\ A(t) > 0 \tag{17}$$

where w_c is the carrier frequency which is considerably greater than the largest frequency component of the modulating signal $A(t)$. $\langle I_i \rangle$ is then analogus to $\langle e_o^2(t) \rangle$, i.e. $1/2|A^2(t)|$ the average being taken over a large number of carrier cycles but over a period over which $A(t)$ does not change appreciably. I_o, on the other hand, is analogus to one half of the ensemble average $\langle A^2(t) \rangle$.

The coherent field amplitude $V_c(t)$, is a slowly varying, zero mean, random process with a normal distribution. The resulting coherent intensity I_c and integrated intensity U_c are as seen earlier exponentially distributed. Thus

$$p(I_c) = \frac{1}{\langle I_c \rangle} e^{-\frac{I_c}{\langle I_c \rangle}} \tag{18}$$

and

$$p(U_c) = \frac{1}{\langle U_c \rangle} e^{-\frac{U_c}{\langle U_c \rangle}} \tag{19}$$

where $\langle I_c \rangle$ and $\langle U_c \rangle$ respectively represent the appropriate ensemble averages. It is assumed here that the random process $V_c(t)$ is stationary in the wide sense and as a consequence $\langle I_c \rangle$ and $\langle U_c \rangle$ are time independent.

Since the correlation time of $V_i(t)$ is very much smaller than the counting period T and since V_c is essentially constant over T the integral

$$\int_t^{t+T} V_c(t')\, V_i(t')\, dt' = 0 \tag{20}$$

and (12) may be written as

$$U = \int_t^{t+T} (V_i{}^2 + V_c{}^2)\, dt' = \int_t^{t+T} I_i\, dt' + \int_t^{t+T} I_c\, dt' \tag{21}$$

i.e.

$$U = \langle I_i \rangle T + I_c T = U_i + U_c \tag{22}$$

Taking ensemble averages on both sides, the total mean integrated intensity is seen to be equal to the sum of the mean integrated intensities of the coherent and the incoherent components.

The mean square intensity may be obtained as follows.

$$V^4 = V_i{}^4 + 4V_i{}^3 V_c + 6V_i{}^2 V_c{}^2 + 4V_i V_c{}^3 + V_c{}^4 \tag{23}$$

Taking ensemble averages on both sides and using the relations:

$$<V_c> = <V_c^3> = 0 \tag{24}$$

$$<V_i^4> = <I_i^2> = I_o^2 + \sigma_i^2 \tag{25}$$

and

$$<V_c^4> = <I_c^2> = 2<I_c>^2 \tag{26}$$

equation (23) may be written as

$$<I^2> = I_o^2 + \sigma_i^2 + 6\ I_o<I_c> + 2<I_c>^2 \tag{27}$$

The normalized second moment, obtained by dividing the second field intensity moment by the square of the average field intensity can be expressed as

$$\frac{<I^2>}{<I>^2} = 1 + \frac{\sigma_i^2 + 4I_o<I_c> + <I_c>^2}{I_o^2 + 2I_o<I_c> + <I_c>^2} \tag{28}$$

In the same manner the normalized third moment of the field intensity is obtained using the relation

$$V^6 = V^6+6V_i^5V_c+15V_i^4V_c^2+20V_i^3+15V_i^2V_c^4+6V_iV_c^5+V_c^6 \tag{29}$$

The resulting expression using the independence of V_i

and V_c and the relation

$$\langle V_c^6 \rangle = \langle I_c^3 \rangle = 6 \langle I_c \rangle^3 \tag{30}$$

is given by

$$\frac{\langle I^3 \rangle}{\langle I \rangle^3} = 1 + \frac{3I_o\sigma_i^2 + 12I_o^2\langle I_c \rangle + 15\sigma_i^2\langle I_c \rangle + 5\langle I_c \rangle^3}{I_o^3 + 3I_o^2\langle I_c \rangle + 3I_o\langle I_c \rangle^2 + \langle I_c \rangle^3} \tag{31}$$

The expressions for the normalized integrated intensity moments are obtained using a similar procedure. They are

$$\frac{\langle U^2 \rangle}{\langle U \rangle^2} = 1 + \frac{\sigma_u^2 + 4U_o\langle U_c \rangle + \langle U_c \rangle^2}{U_o^2 + 2U_o\langle U_c \rangle + \langle U_c \rangle^2} \tag{32}$$

and

$$\frac{\langle U^3 \rangle}{\langle U \rangle^3} = 1 + \frac{3U_o\sigma_u^2 + 12U_o^2\langle U_c \rangle + 15\sigma_u^2\langle U_c \rangle + 5\langle U_c \rangle^3}{U_o^3 + 3U_o^2\langle U_c \rangle + 3U_o\langle U_c \rangle^2 + \langle U_c \rangle^3} \tag{33}$$

If the radiation incident upon the scatterers is thermal radiation, it does not contain any coherent component. No interference effects can be observed using this radiation. The correlation time of the incident radiation and hence that of any component of the scattered radiation is very small compared with the counting period, i.e.

$$\langle U_c \rangle = \langle I_c \rangle = 0 \tag{34}$$

Thus for the scattering of radiation of thermal origin, the expressions for the normalized integrated intensity moments become

$$\frac{\langle U^2 \rangle}{\langle U \rangle^2} = 1 + \frac{\sigma_u^{\,2}}{U_o^{\,2}} \tag{35}$$

and

$$\frac{\langle U^3 \rangle}{\langle U \rangle^3} = 1 + 3\,\frac{\sigma_u^{\,2}}{U_o^{\,2}} \tag{36}$$

It may be seen from equations (1) and (2) that the distribution of the integrated field intensity, p(U), determines the photo-electron counting distribution p(n), through the Poisson transform. If the complete p(n) were known it would be possible to deduce the distribution p(U) by using an inversion procedure[20,21]. Further if p(n,T) were completely known,[20,21] for all T, then it is possible to obtain the distribution of the field intensity I. Practically, however, the determination of p(n,T) for even one value of time presents serious problems. Some of the more important problems encountered are the counter size limitations imposed by total counting times and by the long term stability of the sources of the radiation reaching the detector, the counting errors introduced by the presence of dark current and dynode noise pulses, the requirements of non-zero thresholds and the resulting non-zero threshold fluctuations, the deviation of the actual tube statistics from those predicted from theoretical considerations by Mandel's formula and intensity induced after pulsing caused by residual ionization and by phosphoresence of the photo-cathode and the dynode structure. The last mentioned limitation predisposes the experimentor to using lower intensities and depending upon the quantum efficiency of the photo-cathode material, to relatively small values of average counting rates $\bar{n}$. As a consequence the reliability of the information contained by the higher moments of the counting distribution is poor.

The k th factorial moment of the photocounting distribution is defined by:

$$F(k) = \sum_{n=o}^{\infty} n(n-1)\ldots\ldots(n-k+1)\ p(n). \tag{37}$$

Substituting for p(n) from equation (2) and interchanging the summation and integration this can be expressed as:

$$F(k) = \int_{o}^{\infty} \left[\sum_{n=k}^{\infty} n(n-1)\ldots(n-k+1)\frac{(\alpha U)^n}{n!}\right]e^{-\alpha U}\ p(U)dU \tag{38}$$

Factoring out $(\alpha U)^k$ from the quantity in parenthesis this reduces to:

$$F(k) = \int_0^\infty (\alpha U)^k [\sum_{n=k}^\infty \frac{(\alpha U)^n}{(n-k)!}] e^{-\alpha U} p(U) dU \qquad (39)$$

The quantity in parenthesis in equation (39) is readily identified as $\exp(\alpha U)$ and hence the k th factorial moment of the photo-count distribution can be expressed as:

$$F(k) = \alpha^k \int_0^\infty U^k \; p(U) \; dU$$

$$= \alpha^k <U^k> \qquad (40)$$

Thus the k th moment of the integrated intensity distribution $p(U)$ is seen to be simply related to the k th factorial moment of the photocounting distribution. The presence of α^k, although only as a multiplicative factor imposes further restrictions on the reliability of the values of $<U^k>$ computed.

The parameter α depends upon the area of the detector, the mean spectral sensitivity of the cathode material, pulse-height distribution of the photomultiplier tube and amplifier system and upon the threshold level of the discriminator. The last three factors are subject to variation due to small fluctuations in temperature occuring within the photomultiplier tube and in the associated amplifiers and counting circuitry. All of this adds to the uncertainty in the value of the higher field moments obtained from measurements. Hence, it is reasonable to center attention on the first three moments especially if the quantum efficiency is small. The explicit dependence on α can be eliminated by considering the normalized quantities:

$$\frac{F(k)}{\{F(1)\}^k} = \frac{\langle I^k \rangle}{\langle I \rangle^k} \tag{41}$$

Due to the finite number of samples necessitated by source stability, storage capacity, etc. it becomes necessary to consider the statistical accuracy with which the factorial moments of the counting distributions can be determined. It has been shown[22] that if in a single experiment of M samples, taken such that the inter-sample interval is greater than any coherence time of the field, an unbiased estimate N(k) of the k th factorial moment F(k) is given by:

$$N(k) = (1/M) \sum_{p=1}^{M} n_p (n_p - 1) \ldots . (n_p - k + 1) \tag{42}$$

where n_p denotes the number of counts recorded in the p th sampling period.

The variance of the estimate V(k) can be expressed as:

$$V(k) = \frac{1}{M} \left[\sum_{s=o}^{k} s! \ \binom{k}{s}^2 \ F(2k-s) - \{F(k)\}^2 \right] \tag{43}$$

Since the quantities F(j), j = k, k+1,....,2k are not known, an estimate of the variance may be made using:

$$\text{Est. } V(k) = \frac{1}{M} \left[\sum_{j=o}^{k} j! \ \binom{k}{j}^2 \ N(2k-j) - \{N(k)\}^2 \right] \tag{44}$$

when $\binom{k}{j}$ denotes the bionomial coefficient.

An estimate of the standard deviation of the normalized k th factorial moment $\sigma_N(k)$ may be made by dividing the square root of the above estimate by the k th power of the first moment. Thus

$$\sigma_N(k) = \frac{\sqrt{V(k)}}{\bar{n}^k} \tag{45}$$

where $\bar{n}$ is the first moment or the average number of counts per sample.

From experimental considerations and in order to conserve storage, it is convenient to collect the data as the number of times j counts occurred; j = 0, 1,...,K, where K is the maximum number of counts that can be recorded and K+1 is the channel capacity of the counting system. If for a particular sample the number of counts recorded exceeds K it is convenient to treat that as if K counts had occurred, since, due to the exponential weighting of the Poisson photoelectric detection process, with a negative exponent, the resulting error, especially for small $\bar{n}$, is negligible. For the most part such occurrences are statistically rare anyway. Hence, if $n(j)$; $j = 0, 1, 2, \ldots\ldots, K$ represents the number of times j counts are recorded in an experiment of M samples, the following relations hold.

$$M = \sum_{j=o}^{k} n(j) \tag{46}$$

$$\bar{n} = \frac{1}{M} \sum_{j=1}^{k} j\ n(j) \tag{47}$$

and

$$N(i) = \frac{1}{M} \sum_{j=i}^{k} \frac{j!}{i!}\ n(j);\ i=2,3,.. \tag{48}$$

The rest of the computational procedure is expressed by equations (44) and (45). A FORTRAN Program implementing the above procedure is shown in Appendix B.

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CHAPTER III

DESCRIPTION OF THE SCATTERING SYSTEM

In order to compare the counting distributions of photo-counts obtained due to the scattering of laser and of thermal radiation a scattering system was designed. In this system the aerosols together with the diluting air are made to flow through the scattering volume where they are contained by two sheaths of clean, dry air. The incident radiation travels in a direction perpendicular to the direction of the flow and the scattered radiation is observed in a plane perpendicular to the flow by a photomultiplier which essentially views only the scattering volume. The output of the photomultiplier is processed to yield the desired photo-counting distributions, which at the end of the counting period are printed out on paper tape. The system can be divided into several sub-systems. These may be designated as:

a. Scattering Chamber

b. Aerosol Generation and Flow

c. Light Source and Detection

d. Electronics and Power

A description of each sub-system follows:

Scattering Chamber

The scattering chamber is a box six and one-half feet long, having a cross-section of four feet by four feet. The box is constructed of wood, with the inside coated with a thick coat of non-reflecting flat black paint. At one end of the chamber a laminar flow filter system is connected to the box by means of a black rubber duct. The filtered air from this system enters the chamber through a black screen cloth. This unit provides a laminar flow of clean air through the chamber to remove any dust or other particles. The other end of the box has an open duct feedback to the filter intake so that when the chamber is shut the air inside is re-circulated and filtered by the laminar flow unit. The rubber duct is used to minimize the transmission of vibrations from the filter system blower unit to the chamber walls.

The filter unit used is a class 100 clean air filter. This class filter removes particles 0.3 microns or larger with an efficiency of 99.97% or better, and reduces their density to less than 100 particles per cubic foot. This filter also serves to remove any large particles of the aerosol stream that manage to escape from the containing air streams into the chamber. A photographic view of the exterior of the scattering chamber is shown in

Figure 1 in Appendix C.

Aerosol Generation and Flow

The air flow carrying the aerosol enters the chamber through a pipe placed in the middle and toward the rear of the chamber. The air flows through the pipe in a laminar flow mode for a contained length of three feet. The aerosol stream exits the pipe into the chamber in the form of a free jet stream. This stream is contained by two concentric circular shells of clean dry air which flow from two concentric ring burners placed near the pipe outlet. An inverted funnel is placed above this stream with its stem extending out the top of the chamber where it is connected to a low velocity vacuum suction. The suction serves to scavenge the aerosol stream and the containing air out of the chamber. Figure 2 in Appendix C shows a photograph of the aerosol flow and the ring burner arrangement. It may be noted from the photograph that in the region where the laser light interacts with the aerosol the flow is in the form of a laminar free jet while turbulent mixing of the aerosol stream and the containing air stream takes place well above this region.

The aerosol stream is generated from aqueous solutions of D-sorbitol. This is generated using a fluid atomization aerosol generator purchased from Environmental Research Corporation, and is their model 7300 aerosol generator. The generator produces relatively mono-disperse aerosol particles from aqueous solutions by first generating a poly-disperse aerosol from the solution by fluid atomization, then removing the larger particles from the mixture by an impactor plate. The remaining particles are de-ionized and are then allowed to evaporate. This evaporation process results in a relatively mono-disperse aerosol consisting of particles of the solute and water.

The choice of D-sorbitol for the aerosol generation was made because it readily forms an aqueous solution, and its refractive index is very close to that of water. As far as the refractive index and hence the light scattering is concerned, the resulting aerosol is a two-phase mixture of air particles and particles of D-sorbitol and water. A solution of particles whose refractive index is not close to that of water would form a three-phase mixture with respect to its optical properties. Figure 3 in Appendix C, shows a photograph of the aerosol generator and a diagrammatic representation of the aerosol flow system is shown in Figure 4.

Light Sources and Detection

The light sources used in conjunction with this experiment are a Spectra-Physics model 122 He-Ne laser with a Spectra-Physics model 333, 2.24 cm. beam expander, and a tungsten iodine high intensity lamp bulb with a Sylvania model SG-8 reflector the light from which is passed through a Special Optics monochromatic filter with a 100 Å bandwidth and a 6328 Å center frequency. Both light sources are mounted external to the chamber and the light enters through a hole in the chamber which is at the same height as the top of the pipe carrying the aerosol stream. This entry port is collimated and protected against light leakage. Each of the light beams is collimated so as to have a cross section just large enough to illuminate the entire aerosol stream. In addition the intensity of the laser beam is reduced by the use of a set of polarisers so that in the scattering volume the intensities of both the beams are approximately equal.

The detector used is the EMI model 9502B photomultiplier tube. This tube is mounted in the Electro-Optics Associates model 101 cooling chamber. This detector is mounted on the radial arm of a converted Montgomery Ward radial arm saw. The saw has been removed, and the radial arm is used along with the vertical column and base. The vertical column contains the aerosol stream entry pipe, while the radial arm is connected to a radar mount servomechanism which can be externally controlled to position the radial arm at any angle with respect to the entering light beam and the aerosol column. This system has a positioning accuracy of 0.3 degrees. The detector chamber can thus be positioned at any angular value desired. Total angular rotation of the arm is 185 degrees. The position of the arm is remotely indicated at the instrument panel of the electronic system by means of a synchro system geared to the drive motor for the arm. The drive motor cannot be engaged unless the synchro system is energized, thus eliminating possible reading errors. Stops are placed at each end of the arm traverse in order to insure stoppage of the traverse before the sides of the chamber are reached.

The scattered light enters the cooling chamber and strikes the photo-cathode through a collimating tube. The tube is 15 inches in length and is constructed of black construction paper. The paper is interleaved with aluminum foil to ensure no side radiation entry. At the end of the collimating tube, a polarizing filter is placed in front of the cooling chamber. The collimating tube and polarizing filter can be rotated by multiples of thirty degrees about the axis of the collimating tube. This arrangement allows different polarizations of the scattered radiation to be observed separately.

The photo-multiplier chamber is cooled by passing cold nitrogen gas obtained by evaporating liquid nitrogen through black rubber tubing to the cooling chamber. After cooling the chamber the gas passes out another port and is effused into the chamber. Alternately a flow of dry air cooled by passing it through a long copper coil placed inside a refrigerator can be used in the place of nitrogen. A double wall evacuated glass window is used to thermally insulate the photo-multiplier from the ambient air at the front surface of the cooling chamber. A thermistor mounted at the exit port of the cooling chamber and connected to a Triplett model 850 VTVM is used to monitor the temperature of the tube inside the cooling chamber.

The dynode resistor chain of the photo-multiplier is modified so that the voltage between the cathode and the first dynode is one and one half times the voltage between any two consecutive dynodes. This results in an increased gain for the cathode pulses together with a reduced gain for dynode noise events and permits a better discrimination of the cathode events from the dynode noise events than can be obtained by using a linear dynode chain. The scheme also results in a faster pulse rise time. A μ metal shield is placed around the sides of the photo-multiplier to eliminate the effects of external magnetic fields. The shield is maintained at cathode potential.

Figure 5 of this report is a photograph of the angular positioning system and the photo detector mount assembly. Also shown the two concentric ring burners for the production of the air sheath, and the exit port for the aerosol stream.

Electronics and Power

A block diagram of the electronic system is shown in Figure 6 in Appendix C. The negative voltage pulse generated at the anode as a result of the interaction of the scattered field with the photo-cathode is applied to the input of a two stage pulse amplifier. This amplifier is a Lecroy Research Systems model 133 amplifier which was developed for nuclear event counting. The amplifier has a 200 megahertz bandwidth, and is divided into two stages where each stage has a maximum gain of ten. The amplifier gain is variable in unit steps from one through ten in each stage. Thus the signal can be amplified in caliberated steps from one to one hundred. The output of each stage of the amplifier is of the same polarity as its input and is amplitude limited between plus one half and minus two volts. This limiting is done by the use of fast recovery diodes used in conjunction with linear feedback and is achieved without an appreciable increase in pulse width. The signal cable from the photo-multiplier tube to the input of the amplifier is terminated at both ends. This is done because the output pulses from the photo-multiplier tube vary over a large range of amplitudes and reflections caused by even small mismatches that always exist at coax couplings will otherwise result in false counts and an increased probability of counting a higher number of pulses.

The output of the amplifier is applied through a properly terminated cable to a 100 megahertz amplifier which has a variable threshold. The output of this amplifier is limited by fast recovery diodes to vary between five volts and three volts. The transistors in this amplifier are operated in non-saturating emitter-coupled mode in order to prevent a significant increase in pulse width. The threshold level for this amplifier is provided by a potentiometer controlled voltage. This voltage is set by a potentiometer located on the instrument panel and determines the input threshold at which the output yields a 3.75 volt output level.

The amplifier stage is coupled to a gated emitter coupled logic Schmitt trigger which has 3.75 volt threshold input level. The amplifier and gated Schmitt trigger function as a 100 megahertz, gated, variable threshold, Schmitt trigger circuit with a threshold level range from a minus sixteen millivolts to a minus seven hundred millivolts. In addition the output is conditioned to be compatible with Motorola Emitter Coupled Logic gates (MECL). The gating voltage for this system is obtained from a voltage variable monostable multivibrator circuit whose control voltage is obtained from a potentiometer controlled voltage divider network. The gate is enabled five micro-seconds after the system clock goes positive. The gate width may be varied over both a short range of one hundred nano-seconds to four micro-seconds and a long range of three microseconds to one hundred microseconds. The choice of ranges is determined by a two position switch on the front panel of the instrument board. The gating period determines the period over which photon counting is accomplished.

The Schmitt trigger system output is coupled to the input of a five stage high speed counter. The first stage of the counter is a hundred megahertz J-K flip flop which is followed by three stages of 50 megahertz J-K flip flops. The last stage of the counter is an R-S flip flop. The last stage will be set when the first four stages contain a BCD 9. A one microsecond pulse generated when the clock goes positive resets all five stages of the counter.

When the clock returns to zero, a one microsecond count pulse is generated. The count pulse is supplied to the inputs of ten gates numbered from zero to nine. The outputs of these gates is connected to a set of ten counters which have corresponding numerical designations from zero through nine. The contents of the high speed binary counter will determine which of the gates is enabled. Only one gate is enabled at a time. If for example the contents of the binary counter indicate "three", only gate number three is enabled and a one is added to the contents of counter number three. If the R-S flip flop has been set, then only gate number nine is enabled regardless of the contents of the first four stages of the binary counter. Thus counter number nine will have a one added to its contents whenever the number of pulses in the gating period is nine or more.

The counters are weighted in the following manner: counters "zero" and "one" have five decades each, counters "two" and "three" have four decades each, counters "four" and "five" have three decades each, counters "six" and "seven" have two decades each and counters "eight" and "nine" each have only one decade.

When the count in the most significant decade of any one of the counters reaches nine, the count pulse generating monostable multivibrator is disabled. The disabling of this circuit stops the system from counting any other pulses.

At the same time a light indication on the instrument panel alerts the operator to the condition. This indicates that a single counter has reached its limit. This is done to avoid overflow of any one counter with the consequent loss of information.

The system is equipped with its own 4 kilohertz clock. Over the 250 microsecond period the clock operation is as follows: the clock goes positive (from zero to +5 volts) at the beginning of the period, after 180 microseconds the clock returns to zero until the beginning of the next period. Figure 7 in Appendix C shows the timing diagram for the system.

The content of the counters is printed out sequentially when the control switch is placed in its print position. The print sequence may be described in the following manner. At the start of the print cycle, the print control counter is reset to zero, this also connects the counter number zero to the input of the printer. After a twenty microsecond delay a print command pulse is generated. This command pulse causes the printer to print the contents of the three most significant decades of the "zero" counter. The digit stored in the control counter, namely "zero", is also printed alongside for identification purposes. At the end of the print cycle a contact switch is lifted from ground within the printer. This causes the print control counter to advance by one and after a delay of 100 milliseconds another print pulse is generated which causes the printing of the contents of the three most significant decades of counter number one together with "one" the digit stored in the print control counter. The sequence is repeated until the contents of the three most significant decades of the counters "two" through "five" are printed. At the next print cycle the contents of the counter "six" are printed, followed by the content of the least significant decade of counter "two". This is followed by the printing of the contents of counter "seven" together with those of the least significant decade of counter "three" at the following print cycle. Together with the contents of the "eight" counter are printed the contents of the two least significant decades of counter "zero" and the two least significant decades of counter "one" are printed besides the contents of counter number nine. At the next lifting of the contact switch within the printer the print control counter is reset to zero and the cycle is again initiated. The printing ceases when the control switch is returned to the off position. A sample of the format together with a listing of the contents of the different counters is presented in Figure 8 in Appendix C. The adoption of this format which is a little difficult to interpret results in considerable savings in the amount of decoding circuitry necessary for the sequential printing.

The ten counters which are storing the probability distribution of the number of counts during the period set by the gate are reset simultaneously by pressing a reset switch on the front panel. This insures that the only modification to the information in the counters is a complete erasure. Figure 9 is a photograph of the front panel of the electronics package. In this figure the printer, which is a modified Beckman model 1453 printer, may be seen on top of the assembly, while the Lecroy Research System pulse amplifier and the power supplies may be observed in the lower portion of the instrument rack. Figure 10 is a photograph of the integrated circuits, amplifiers, triggers and gates previously mentioned.

Power for the system is supplied from a Sorenson model ACR 2000 line voltage stabilizer. The D. C. power supplies utilized for the electronics and the photomultiplier tube include a Kepco model SC-38-2M (0-38 volts, 2 amperes), a Harrison Laboratories model 6384A integrated circuit power supply (4-5.5 volts, 0-8 amperes), a Harrison Laboratories model 6205 dual D. C. power supply (0-40 volts, 0-0.3 amperes), and a Harrison Laboratories model 6516A high voltage power supply (0-3 kilovolts, 0-6 milliamperes). The power supply voltage levels used in the system are: +5 volts, + 12 volts, + 24 volts and -12 volts.

CHAPTER IV

EXPERIMENTAL PROCEDURES

The photomultiplier tube was placed in the cooling assembly to which the collimating tube was attached and a cap was placed on the front end of the collimating tube in order to ensure that no light entered the photomultiplier tube. High voltage was applied and the tube was run in the dark for a period of seventy two hours during which an analysis of the dark current was made. The pulses from the photomultiplier tube were amplified by the Lecroy Research System Model 133 amplifier and were applied to the input of a Fairchild Model 766 H/F oscilloscope. Care was taken to ensure that the cable connecting the output of the photomultiplier tube to the input of the amplifier was properly terminated at both ends and the cable connecting the output of the amplifier to the oscilloscope was also properly terminated into its characteristic impedance. The reflection coefficients at each of these terminations were made less than 0.02. The dark pulses were observed on the oscilloscope to ensure that there was no appreciable double pulsing in the tube. Double pulsing which is due to the ionization of residual gas molecules, if any, in the tube would indicate that the tube is unsuitable for use in photocounting experiments. Of the three tubes tested only one did not show any significant double pulsing.

The pulses from the output of the Lecroy Research System (LRS) amplifier were next applied to the input of the General Radio, Model GR 1153-AP counter which was operated at its lowest threshold setting of 100 mV. Dark current counts were integrated for periods up to 10 seconds and recorded for different gain settings of the LRS amplifier and a plot of the logarithm of the mean count rate vs an equivalent threshold, obtained by dividing 100 mV by the gain setting of the amplifier was made. This was done to ensure that dynode noise pulses did not dominate the lower end of the pulse height spectrum of the photomultiplier tube. During this time the tube temperature was maintained close to room temperature by passing dry air at room temperature through the cooling chamber assembly.

The mean dark current was next obtained for different high voltage settings, keeping the amplifier gain at 100 and a plot of the logarithm of the dark current count vs the logarithm of the voltage setting, shown in Figure 1 in Appendix A, was made. This was compared to a similar plot, of the logarithm of the photomultiplier tube gain vs the logarithm of the applied voltage, which was supplied by the manufacturer. From this comparison the optimum voltage to operate the tube was ascertained. A voltage just below the knee of the dark count vs voltage curve was chosen. The choice was verified by making a plot of the logarithm of the number of dark current counts per second vs the equivalent threshold for this voltage and for a higher voltage. Whereas at this voltage the plot was a straight line, it was found that at the higher voltage the plot tended to deviate upwards at small threshold settings, indicating that at the higher voltage setting noise pulses from the first dynode when amplified through the dynode system became prominent when compared with the cathode event pulses.

A flashlight bulb was then placed in front of the photomultiplier tube and connected with a variable, regulated, d.c. power supply. The light from the bulb was passed through a polarizer and an interference filter before illuminating the tube. The voltage supply to the bulb was slowly increased while monitoring the counts on the General Radio counter until approximately one hundred thousand counts per second were registered. A curve of the logarithm of the mean count rate vs equivalent threshold voltage level was then plotted. This curve, shown in Figure 2 in Appendix A, is almost a straight line indicating that the tube was linear at this high counting rate. The slight decrease in the number of low height pulses results from dead time effects which begin to become effective at these counting rates.

The tube was run with the light on for several days. Using the system described in the previous chapter and a counting period of 9.4 micro-seconds, counting distributions were obtained periodically from which the field distribution moments, the normalized moments and the variances were computed as outlined in Chapter II. These were compared with those calculated for the Poissson distribution. After several attempts the distribution became very close to the Poisson distribution. Figure 3 in Appendix A shows two samples of the distribution and the corresponding factorial moments, normalized factorial moments, and their variances obtained at this time.

It was observed that the distribution approached a Poisson distribution after the light was turned on for over 48 hours. The phenomenon may be attributed to the cleaning and stabilization of the dynodes with the passage of current through them. Thermal stabilization may also be a possible factor. In order to ensure proper operation, a light emitting diode was placed in the collimating tube, such that although it could be seen by the photomultiplier tube, it would not obstruct the path of light entering the phototube from the front end of the collimating tube. The light emitting diode was supplied by a variable, regulated d.c. current and was always turned on, so that the tube was continuously counting at approximately two hundred thousand counts per second, whenever the system was not being used to measure the scattered light.

When the system was used to make measurements, the diode current was reduced to make the diode photo-electron counting rate approximately equal to the average signal counting rate. The diode current was turned off, using a switch on the front panel, while counting from the scattered radiation. Periodically, the beam of light incident on the scatterers was obstructed, and the light emitting diode was turned on and photo-electron distributions due to its radiation were recorded. These served as a check on the stability of the counting system and as a reference counting distribution with respect to which all scattering data could be related.

Two sets of aerosols were used in the experiment. The first one was a D-Sorbitol aerosol made from an aqueous solution by atomizing the liquid and evaporating the resulting aerosol to yield fine, uniform spherical particles. The choice of D-Sorbitol was made, as explained earlier, because the resultant aerosol, as far as scattering is concerned, is a one phase mixture due to the closeness of the refractive index of D-Sorbitol to that of water vapour particles. One problem that resulted was that the intensity of the scattered radiation was very weak and resulted in measurements with poor reliability. As an additional check on the validity of the measurements, another aerosol obtained by atomizing liquid Dioctyl Phthalate (DOP) in a Royco Instruments, Inc. Model 258, smoke generator was used which yielded larger particles causing higher intensities of the scattered radiation. As no apparatus was available for determining the particle size distribution or their flow rate, these were estimated from the data supplied by the manufacturers of the aerosol generators used. In the case of the sorbitol aerosol the mean particle size (number median) was estimated at 0.1 micron with a geometrical standard deviation of 1.6. In the case of the DOP aerosol the mean particle size was estimated to be 0.55 micron with a geometrical standard deviation of 1.35. In the case of the sorbitol, which was generated from a 5% solution by weight of sorbitol, the flow rate and the mean number of particles were estimated by using the calibration curve of the aerosol generator and by recording the rate of consumption of the solution. For the settings used, the flow rate was found to be 22.5 litres per minute, the solution consumption rate was 9.8 ml. per hour. The resulting average particle velocity was calculated to be 1.36 meters per second. The mass median of the sorbitol aerosol was approximately 0.3 microns i.e. the average volume of the particles was approximately 2×10^{-14} c.c. The average number of particles per c.c. of air flow calculates to approximately 2.2×10^{7}. In order to ensure uniformity of the aerosol, about 2000 c.c. of sorbitol solution was prepared at the beginning of the experiment.

For lack of measuring equipment no estimate could be made about the mean velocity of the DOP aerosol. Considering the fact that only one jet of the aerosol generator was used and that even this jet was opened approximately one fifth of the full scale opening, together with the fact that a lower pressure was used to form the aerosol stream the DOP aerosol stream had a velocity smaller than that of the sorbitol aerosol stream. Furthermore, even with a larger particle size and the resulting aerosol having a scattering cross section that was considerably larger than that of the sorbitol stream, the rate of fluid consumption was of the same order as that of the 5% sorbitol solution. It may therefore be reasonable to state that the mean velocity of the DOP aerosol stream was less than 1 meter per second.

The average gain of the tube was estimated using the data supplied by the manufacturer. The cathode sensitivity for the particular tube was specified as 66 micro-amperes per lumen (at the peak luminous sensitivity) and the anode sensitivity at the operating voltage of 1580 volts was measured by the manufacturer as 2000 amperes per lumen (also at the peak luminous sensitivity). The average gain of the tube is therefore 3×10^7.

A typical data run may be described in the following manner. The laser was turned on and allowed to stabilize for a period of one hour during which time the air drying column was filled with a fresh charge of silica gel and reconnected to the system and the old charge was heated in an oven in order to regenerate the dessicant. The room lights were turned off, the scattering chamber was opened and the cap that was placed on the end of the collimating tube was removed. The chamber was next closed and the laminar flow unit was turned on in order to clean up any dust particles in the chamber.

At the end of the warm up period, the vacuum suction system was turned on which was followed by turning on the air supply to the aerosol generator to start the aerosol stream. The current to the light emitting diode was then turned off, the cover on the light entry port was removed and the angle at which the scattered radiation was observed was reduced to approximately 16 degrees. At this position the signal was usually maximum and no direct radiation of the source entered the collimating tube. The resulting counting rate on the General Radio counter, which was connected to the output of the LRS amplifier, was noted for 20 samples, each of one second duration. The thermal source was then placed in the beam path and current was supplied to it and the narrow band filter and the polarizer were placed in mounts provided for them on the entrance port of the scattering chamber. The placement of the thermal source obstructed the laser beam so that its radiation could no longer enter the chamber. The intensity of the thermal source was adjusted using a rheostat until the scattered thermal radiation produced approximately the same counting rate as that produced by the scattered laser radiation. The power to the thermal source was then turned off, the entrance port to the chamber was covered, the l.e.d. was turned on, and using the potentiometer on the instrumentation panel, the voltage to the circuit that controlled the current to the l.e.d. was adjusted until the counting rate due to the light emitted by the diode was approximately equal to the rate due to the scattered laser radiation. The General Radio counter was then disconnected, and the output of the LRS amplifier was connected to the input of the counting system and counting distributions for the l.e.d. light were recorded. After a set of counting distributions, usually five in number, the l.e.d. was turned off, the thermal light source was turned on and a set of several counting distributions, usually containing between five and twenty distributions was recorded. The thermal source, the filter and the polarizer were then removed and a similar set of counting distributions for the scattered laser radiation was recorded. Finally, the cover was again placed on the port of the scattering chamber and the l.e.d. was again turned on in order to record a set of counting distributions from its radiation. The system parameters i.e. the scattering angle or the counting period were then changed. If this resulted in a change in the intensity of the scattered radiation, the light emitted from the l.e.d. was correspondingly adjusted and sets of data as described previously were taken. As a check on the system stability some of the afore-mentioned sets were taken another time towards the end of the run. The interleaving of the various sets of data was done in order to ensure that long term drift if any would not jeopardize the validity of the comparison.

At the end of the run, the air supply to the aerosol generator was turned off, the scattering chamber was opened, the cap was placed on the end of the collimating tube and the chamber was reclosed. The room lights could now be turned on and the rest of the system was then shut down except for the high voltage to the photomultiplier tube, the cooling air flow and the current to the light emitting diode. The General Radio counter was again connected to the output of the LRS amplifier and the light output of the diode was adjusted to give a counting rate of approximately 200,000 counts/second.

The data in every channel from all of the counting distributions in each set were added together to obtain a sum of the counts in that channel. The total data was then processed in the manner indicated in Chapter II to obtain the average factorial moments and normalized factorial moments of that set of samples. The computed normalized standard deviations of several sets were plotted against the average counting rate per second. It was observed that the standard deviations, of sets of the same size and counting periods, were inversely proportional to the counting rate as long as the latter was less than approximately 100,000 counts per second. For higher counting rates the variances tended to level off to constant values. This may be caused by the increase in variations of the distributions caused by dead time effects. Accordingly, when polynomial regression was performed on the data sets, the data points were weighted in a manner inverse to the relationship held by the standard deviations.

The plots of the experimental data may be observed in Appendix A. Also shown on the figures in Appendix A are the curves obtained by first and second order polynomial regression through the data points. A discussion of the results follows in the next chapter.

CHAPTER V

RESULTS AND CONCLUSIONS

The nature of the radiation scattered from a flow of airborne aerosols was investigated experimentally by observing the statistics of the photo-electron counting distributions obtained by allowing the scattered radiation to be incident on a photomultiplier tube. Moments of the distributions of the integrated field intensities of the radiation incident on the photomultiplier tube were computed by considering the factorial moments of the photo-electron counting distributions obtained from experimental measurements, these were normalized with respect to the corresponding mean counting rates, i.e. the first moments. The normalized moments for the several experimental situations are shown in Figures 4 thru 11 in Appendix A.

The main goal of this investigation was to develop a system to obtain photo-electron counting distributions for scattered radiation and to determine whether any differences could be observed between the counting distributions obtained for scattered laser light and for scattered incoherent thermal light of the same mean frequency. This has been accomplished as may be observed in Figures 4 thru 7 of Appendix A. Figures 4 and 5 show the normalized second and third factorial moments of the counting distributions obtained from laser and thermal radiations scattered from a sorbitol aerosol versus the average number of counts per sampling interval. Also shown are the same moments for light emitted from the light emitting diode. Similar curves for light scattered from DOP are shown in Figures 6 and 7. It may be remarked that for the scattering from sorbitol the scattering angle and hence the intensity were kept constant at 16.2 degrees and the variation in $\bar{n}$ resulted from a variation in the sampling period. In the case of scattering from DOP, the sampling period was kept constant at 3 micro-seconds and the variation in $\bar{n}$ resulted from a variation in the angle of observation and the consequent variation in intensity. It is important to note that in all of these cases the data from the laser light scatter and the light emitting diode and for thermal light scatter and the light emitting diode was obtained in a sandwiched manner so that there was no possibility for any long term drift to influence the comparison. The scattered laser radiation produced second order moments that were consistently larger than those produced by the scattered thermal radiation. Both of these were significantly higher than those produced by light emitted from the light emitting diode. With increasing values of $\bar{n}$, curves plotted from data samples for the normalized second factorial moments extrapolate approximately to 1.0 for both the scattered thermal radiation and for the radiation from the light emitting diode. In the case of the scattered laser radiation they extrapolate to values of approximately 1.02 for the sorbitol aerosol and 1.03 for the DOP aerosol. The fact that the curves for the scattered laser radiation do not extrapolate to a value close to 2.0 indicates that the simpler models developed in Chapter II are not applicable.

In order to observe the applicability of the model described in Chapter II, the following factors may be considered. The mean counts per sample, i.e. $\bar{n}$, is proportional to the integrated field intensity U, which for a constant counting period results due to increasing values of I_o and for a constant I_o results due to increasing T values. Referring to Equation (32) in Chapter II, upon dividing both numerator and denominator of the second factor on the right hand side by U_o^2, and defining α and β by;

$$\alpha = \frac{\sigma_u}{U_o} \tag{49}$$

and

$$\beta = \frac{\langle U_c \rangle}{U_o} \tag{50}$$

the following result may be obtained

$$\frac{\langle U^2\rangle}{\langle U\rangle^2} = 1 + \frac{\alpha^2 + 4\beta + \beta^2}{1 + 2\beta + \beta^2} \qquad (51)$$

In the case of scatter from sorbitol where the scattered radiation is the same for all the data points it is easy to see that the parameter α decreases with increasing T. This is due to the fact that σ_u^2 is directly proportional to σ_I^2 and inversely proportional to T, the latter resulting as a consequence of smoothing, due to integration, of both the variance of the integrated intensity and the increase in the second moment caused by threshold fluctuations. The parameter β also decreases due to the process of integration. It has been show that[23,24] as the ratio of the counting period to the correlation period of the radiation increases, the normalized second moment will decrease. This may be interpreted to imply that the ratio of the coherent component to the incoherent component will also decrease. The net result is that with increasing $\bar{n}$, the curves for the normalized second factorial moment all show a decrease and the curve denoting scattered laser radiation and the curve denoting scattered thermal radiation come closer to one another. Also in the limit the latter approaches a value of 1.0 while the former approaches a slightly higher value.

In the case of scattering from DOP, where the counting period T is constant, the reduction in σ_u and the consequent reduction in the parameter α are attributed to the following:

1. With increased intensity there is a reduction in that part of the σ_u^2 that results from threshold variations.
2. Dead time effects become more prominent because of the increase in the average number of photo-electrons emitted from the cathode.
3. Any contributions caused by high level dynode noise pulses become less effective due to increased signal to noise ratios, so do variances arising from shot noise and quantum noise.

The curves for both the scattered thermal radiation and the radiation from the light emitting diode decrease towards a value of 1.0 as $\bar{n}$ increases. In this case however, the difference in the normalized second moments of the scattered laser and thermal radiations does not change as significantly as in the previous case. From Equation (3) this difference can be expressed as:

$$\frac{\langle U^2\rangle}{\langle U\rangle^2} - \frac{\langle U^2\rangle}{\langle U\rangle^2}\bigg|_{\beta=o} = \frac{\alpha^2 + 4\beta + \beta^2}{1 + 2\beta + \beta^2} - \alpha^2$$

$$= \frac{2\beta + \beta(\beta+2)(1-\alpha^2)}{1 + 2\beta + \beta^2} \qquad (52)$$

As α^2 and β^2 are small compared with 1, the above difference can be expressed as:

$$\frac{\langle U^2 \rangle}{\langle U \rangle^2} - \frac{\langle U^2 \rangle}{\langle U \rangle^2}\bigg|_{\beta=0} \simeq \frac{4\beta}{1 + 2\beta} \qquad (53)$$

In this case where T is constant and I_o is changing, the parameter β can be expected to remain constant as long as no part of the incident radiation enters the collimation tube; i.e. as long as all the radiation reaching the detector has been scattered from the aerosol. The curves for the normalized third factorial moments show similar trends but due to the increased effects of experimental errors in this case, it is not possible to discuss their detailed structure.

Figures 8 thru 11 show the normalized second and third factorial moments of the scattered laser radiation and radiation from the light emitting diode versus the inverse of the average counting rate per unit time with the sample period as a parameter. Since, in this case, no direct comparison between scatter from thermal radiation and laser radiation was to be made, the intensity of the incident radiation could be increased considerably. The attainment of larger values of collimated intensity of the thermal radiation was a serious limitation in the earlier set. A less dense aerosol was chosen with an intention of reducing multiple scatter and observing the behaviour of the system. In this instant both the angle of observation and the sampling period were varied. The data for the 3 micro-seconds from Figures 6 and 7 have been redrawn into this set to provide a comparison. The dependence on the average counting rate and the sampling period can be seen more clearly in this set.

The data shown in Figures 8 thru 11 were regressed using polynomial regression with a weighted quadratic cost factor. The weighting function used has been described in the previous chapter. The results of the regression are shown in Tables 1 and 2. Since the data for the 3 micro-second counting period is from a different aerosol concentration than that for the rest of the counting periods, the corresponding polynomial values are omitted from the tables. The parameter k in the expressions in the tables is inversely proportional to the average counting rate and is therefore proportional to $1/\langle I\rangle$ where $\langle I\rangle$ represents the average field intensity on the cathode surface; therefore, the normalized second and third factorial moments of the integrated field intensity can be expressed by:

$$\frac{\langle U^2\rangle}{\langle U\rangle^2} = 1 + a + \frac{b}{\langle I\rangle} + \frac{c}{\langle I\rangle^2} \tag{54}$$

TABLE 1

SECOND ORDER POLYNOMIAL REGRESSIONS

NORMALIZED SECOND FACTORIAL MOMENTS

k = 1/ Average counts per micro-second

COUNTING PERIOD	REGRESSED POLYNOMIAL
SCATTERED LASER RADIATION	
2 Sec.	$1.0618 + 0.0020\ k + 0.00038\ k^2$
5 Sec.	$1.0602 + 0.0052\ k$
10 Sec.	$1.0575 + 0.0040\ k$
20 Sec.	$1.0510 + 0.0028\ k$
RADIATION FROM LIGHT EMITTING DIODE	
2 Sec.	$1.0116 + 0.0024\ k + 0.00028\ k^2$
5 Sec.	$1.0138 + 0.0027\ k$
10 Sec.	$1.0135 + 0.0020\ k$
20 Sec.	$1.0110 + 0.0014\ k$

TABLE 2

SECOND ORDER POLYNOMIAL REGRESSIONS

NORMALIZED THIRD FACTORIAL MOMENTS

$k = 1/$ Average counts per micro-second

COUNTING PERIOD	REGRESSED POLYNOMIAL
	SCATTERED LASER RADIATION
2 Sec.	$1.127 + 0.0166\ k + 0.0041\ k^2$
5 Sec.	$1.108 + 0.0252\ k + 0.0003\ k^2$
10 Sec.	$1.1126 + 0.016\ k + 0.0002\ k^2$
20 Sec.	$1.1055 + 0.004\ k + 0.0003\ k^2$
	RADIATION FROM LIGHT EMITTING DIODE
2 Sec.	$1.0318 + 0.0044\ k + 0.0029\ k^2$
5 Sec.	$1.0037 + 0.0170\ k$
10 Sec.	$1.0005 + 0.0095\ k$
20 Sec.	$1.000 + 0.0066\ k$

and

$$\frac{\langle U^3 \rangle}{\langle U \rangle^3} = 1 + a' + \frac{b'}{\langle I \rangle} + \frac{c'}{\langle I \rangle^2} \tag{55}$$

where the coefficients a, b, c, a', b', and c' depend upon the counting period T.

From the expressions in the tables, the following observations may be made. The parameter a in Equation (54) is almost time independent. The small differences seen can be accounted for as experimental errors and errors due to regression. This parameter does depend, however, on the nature of the radiation incident on the photo-cathode. The departures from the value 1.0, i.e. from the theoretical values expected for a Poisson distribution, for the third and second factorial moments have a ratio of about two for the scattered laser radiation, which is lower than a value of three or more that is predicted from the theoretical expressions developed in Chapter II. This effect is caused by dead time[25], the random (poisson distributed) photo-electrons due to phosphorescence being a contributing factor. The effect is even more prominent in the case of the radiation from the light emitting diode where the resulting reduction in the third factorial moment at the higher intensities more than compensates for the expected increased variance due to threshold fluctuations and ion produced afterpulses.

The parameters b and b' from Equations (54) and (55), which denote the proportionality inverse to the average counting rate show an increase between the counting periods of two and five micro-seconds, and then decrease with increasing counting periods. The rate of decrease is larger for the scattered laser radiation than for the radiation from the light emitting diode. The behaviour of the normalized factorial moments with an inverse relationship with the average intensity is the result of several effects. Shot noise, threshold fluctuations, and dynode fatigue[26] all give rise to an increased variance with a dependence inversely proportional to the average intensity of the scattered radiation. Moreover, smoothing due to integration which results with increasing sample times accounts for the 1/T behaviour of this increase. Pulses due to radioactive isotopes present in the glass envelopes[27], gamma rays and cosmic particles all produce decrease in the signal to noise ratio which has a $1/\langle I \rangle$ dependence. In general the latter make a small contribution that is independent of time but the quantization errors introduced will have a 1/T behaviour. The higher rate of decrease of the parameters b and b' for the scattered laser radiation compared with those for the light emitting diode results due to an additional contribution caused by the integration of the coherent component and a small contribution due to fluctuations in the number of particles and in the source intensity. No definite mechanisms for the reduction in these parameters for small counting periods are known and the observed effect is probably due to the regression and to the approximations in the weighting factors used. Another possible factor is the effect of quantization caused by the reduction in the probability of counts in the higher channels due to dead time.

The $1/\langle I\rangle^2$ dependence represented by the parameters c and c' is produced by a reduction in the signal to noise ratios at the lower intensities due to the $1/h^2$ noise[28] and due to the quantization effects caused by dark current pulses having a non-Poissonian nature that have been observed[29] in EMI 9502B tubes. In the case of scattered laser light a small contribution is also made by the factor β^2 of Equation (51). The reduction of these factors for the larger counting periods is evidently due to the quadratic effects of time integration of these factors.

The errors caused by the limitations of the experimental apparatus such as counting capacity, source stability, stability of the flow of scatterers and temperature stability impose a limit on the reliability of the information obtained. One major problem is the low quantum efficiency and the relatively slow response of the EMI 9502 B photomultiplier tube, the selection of which was governed by economic considerations. The performance of the tube was found to be otherwise quite satisfactory. The rest of the electronic equipment including the LRS amplifier performed very well. A small amplitude of some 1 MHz noise that was present in the A.C. power line did couple through to the high voltage supply due to imperfect filtering by the H.P. high voltage power supply. The amplitude of this spurious signal on the line was small and a two section RC filter introduced in series with the high voltage line remedied this situation.

One particular phenomenon that has not been considered in the above discussions is the effect of Brownian motion on the ratio β defined in Equation (50). In Chapter II it was shown that the average velocity of one micron particles, having a unity specific gravity, produces a doppler spread of approximately 6 kHz. For the case of the sorbitol aerosol the size of the particle with the average mass is approximately 0.3 microns and the doppler shift resulting from the average velocity of the particles due to Brownian motion will be slightly larger than 38.2 kHz. Due to the Maxwellian distribution[30] of the Brownian velocities a significant percentage of the scatterers will induce a doppler shift above this frequency, reducing the correlation period of the radiation. Recently Germogenova and Siegel[31] have studied the effect of the spread due to Brownian motion and have concluded that there is a loss of coherence even in a forward transmitted beam. The extension of this to the scattered radiation is only natural. As noted earlier, the net effect of this is to reduce the ratio of the coherent component to the incoherent component of the scattered radiation. In the case of the DOP aerosol the size of particles having the average mass exceeds 1 micron and the previously mentioned effect will be less pronounced. As no further information on the size and mass distribution of the aerosols was obtainable, a detailed analysis of the contributions of the Brownian motion could not be made.

The preceding discussions show that the model constructed in Chapter II is a valid one and the integrated field intensity of the scattered field consists of a sum of two factors; a near constant, incoherent component with a narrow distribution together with a coherent component resulting from the interference effects in the scattered radiation. Experimental evidence indicates that the nature of the field scattered by aerosols from an incident coherent field is different from that of the field scattered from an incident incoherent field of thermal origin. Further it is shown that the former field does induce correlated counting distributions in accordance with the expected behaviour of the photomultiplier detector used, and that photon counting is a useful tool in studying the scattering from aerosols and other random media since it offers an additional dimension of information.

A major contribution of this research effort was the development of a system that can be used to study angular distribution of scattered radiation, both from the standpoint of making intensity measurements and photo-electron counting measurements, from aerosols and other random media. The electronics have been designed to make the system versatile. A substitution of two printed circuit cards and the addition of an external synch pulse can convert this system into a signal amplitude distribution analyser with a sampling rate capability in excess of 5 MHz. By disabling the system clock and counting pulse, each of which can be accomplished by pulling out one transistor from its socket, the system can be modified for the analysis of event time distributions with arbitrary sampling periods and with adjustable noise immunity. The only limitations of this system are exponential channel weighting, limited counting capacity and a very small number of channels, all of which are a result of budgetary limitations. Another tedious problem is the requirement of manual transfer of data from printer output to computer cards for further processing. The system does have a built in capability for overcoming these limitations with little modification and considering all factors is a fairly good prototype for a larger system.

This investigation has provided for the experimental confirmation of the existance of interference effects in radiation scattered from aerosols, and has shown that for coherent incident radiation some correlation does exist between the incident and the scattered radiation. The natural extension of this is to develop techniques for the recovery of this information using amplitude modulated incident radiation. Several other problems related to the present investigation can be suggested. Due to the limited experimental accuracies the question of variation of co-rrelations in the scattered radiation with angular position could not be studied and should be further explored. Another aspect is the study of correlations with variation in aerosol size and composition. Yet another aspect is the photo-electron counting distributions of modulated signals, a study that is presently being undertaken. Associated theoretical investigations include the effects of Brownian motion on the correlation periods of radiation scattered from aerosols of known size distributions, the effects of turbulence on the coherence properties of scattered radiation and the recently suggested[32] increase in coherence of the scattered radiation by propagation as suggested by the Van Cittert-Zernike theorem[33] and its experimental verification.

The increase in the variance of the scattered coherent radiation over that of the scattered thermal radiation suggests that this effect will cause errors in the estimation of the properties of aerosols, especially the number distribution, from observations of the scattered radiation. For example, if the aerosols were identical, the distribution of the scattered intensity for incident thermal radiation will be the same as the distribution of the number of aerosols over the observation period. Incident coherent radiation, however, will give rise to a different intensity distribution due to interference effects. Some errors in the interpretation of lidar returns and in the solution of the associated inversion problem can also be attributed to this phenomenon and its better understanding will provide for the estimation and compensation of these effects.

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APPENDIX A

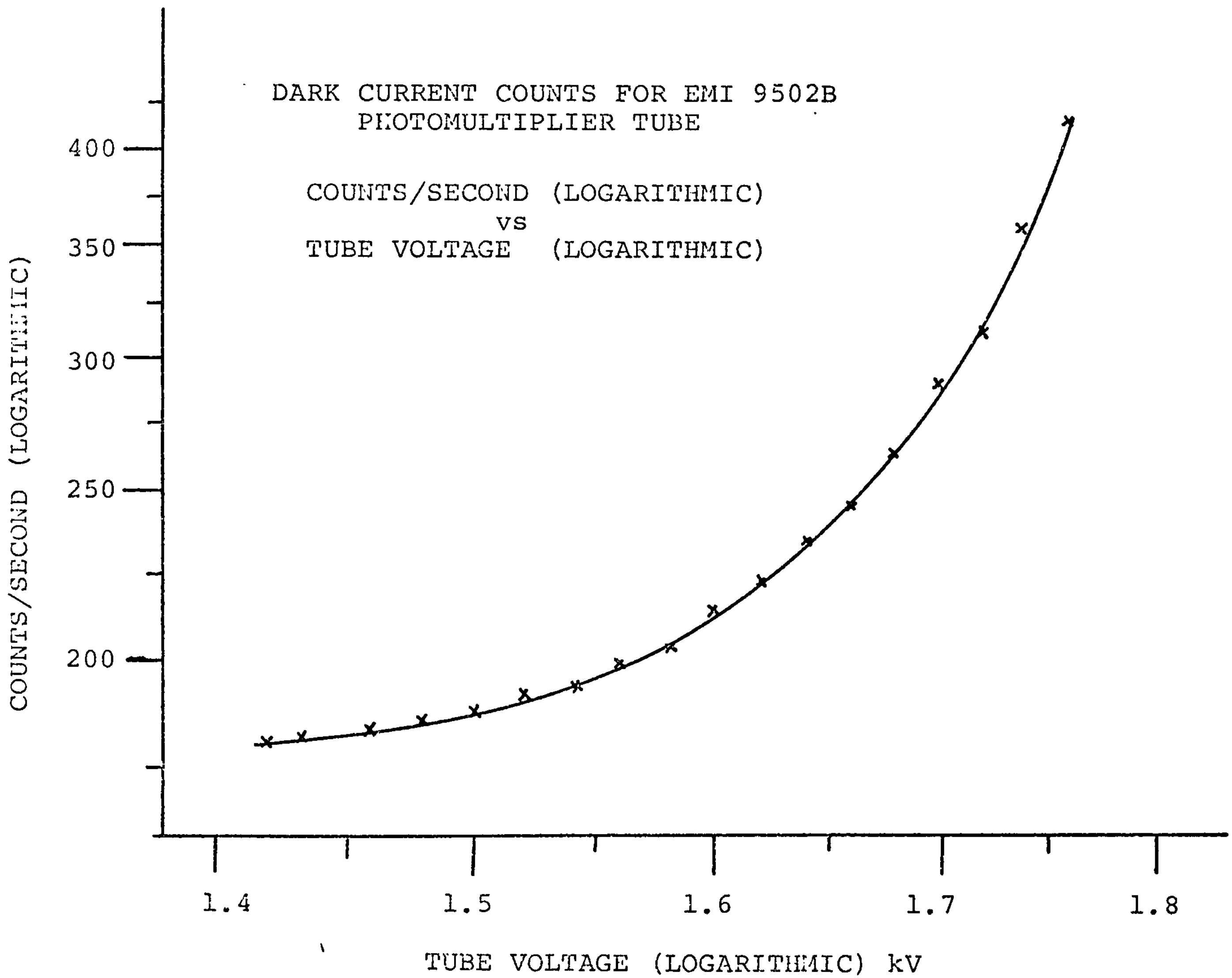
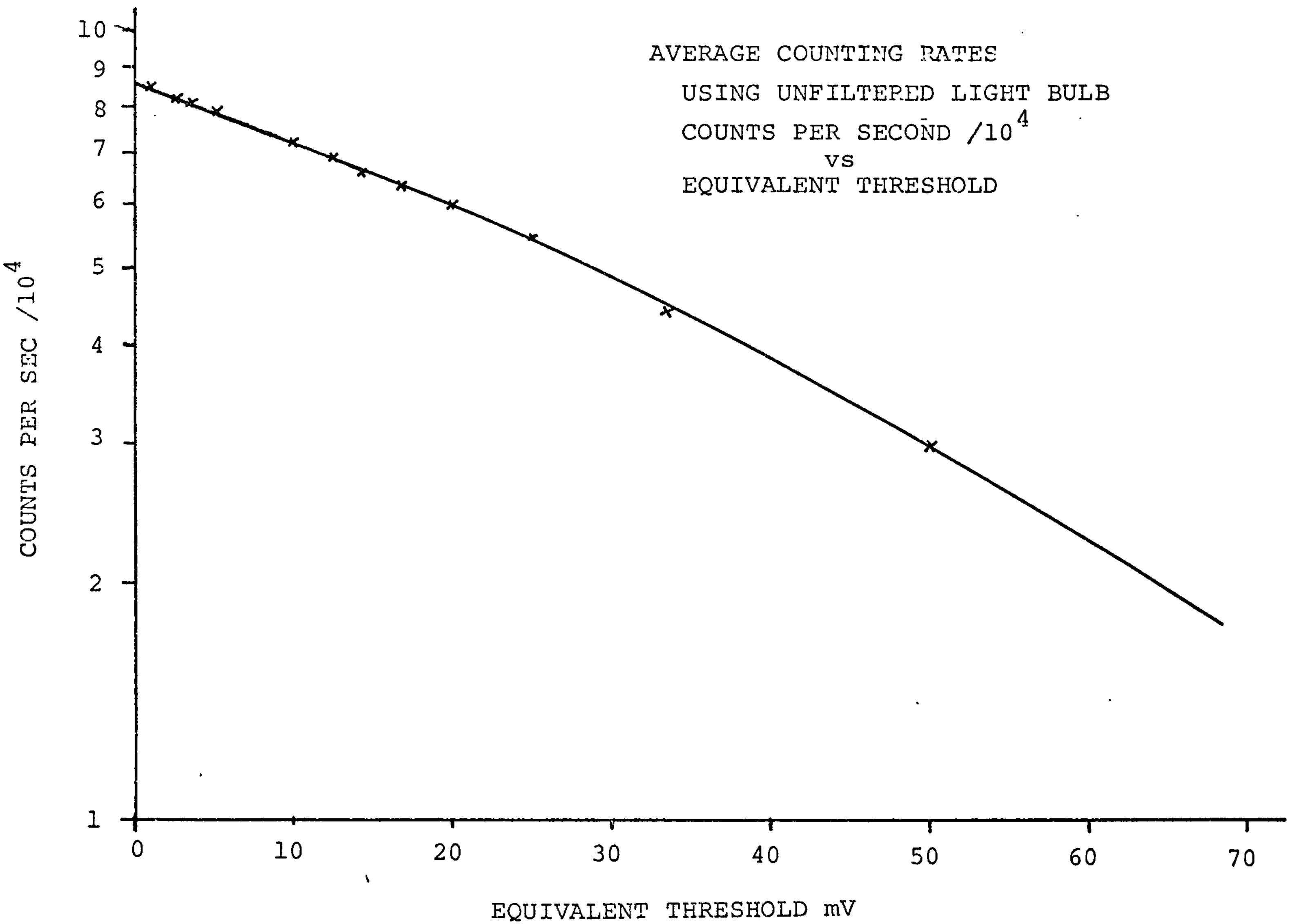

DARK CURRENT COUNTS FOR EMI 9502B
PHOTOMULTIPLIER TUBE
COUNTS/SECOND (LOGARITHMIC)
vs
TUBE VOLTAGE (LOGARITHMIC)
400
350
300
250
200
COUNTS/SECOND (LOGARITHMIC)
1.4
1.5
1.6
1.7
1.8
TUBE VOLTAGE (LOGARITHMIC) kV

FIGURE 1

FIGURE 2



AVERAGE COUNTING RATES
USING UNFILTERED LIGHT BULB
COUNTS PER SECOND /10^4
vs
EQUIVALENT THRESHOLD
COUNTS PER SEC /10^4
EQUIVALENT THRESHOLD mV
10
9
8
7
6
5
4
3
2
1
0
10
20
30
40
50
60
70

TUBE TEST TAKEN BEFORE THE BEGINNING OF THE EXPERIMENT
DATA TAKEN WITH POLARIZED NARROWBAND RED LIGHT

Counting Period : 9.4 Micro-seconds

Data Recorded by the Printer

Sample #1	Sample #2
9083	9003
8079	8070
7004	7008
6020	6100
5024	5115
4221	4528
3162	3255
2900	2900
1374	1220
0735	0263

Corresponding Recording on Computer Cards

Channel No.	Sample #1	Sample #2
9	0	0
8	0	0
7	0	0
6	2	10
5	24	115
4	221	528
3	1624	2558
2	9000	9000
1	37483	22003
0	73579	26370

Data Computed Using the Program

Normalized Factorial Moment	Sample #1	Sample #2
Second	1.0015	1.0075
Third	1.0760	1.0341
Fourth	1.1379	1.0344
Fifth	1.0969	0.8679
$\bar{n}$	0.53317	0.8323

FIGURE 3

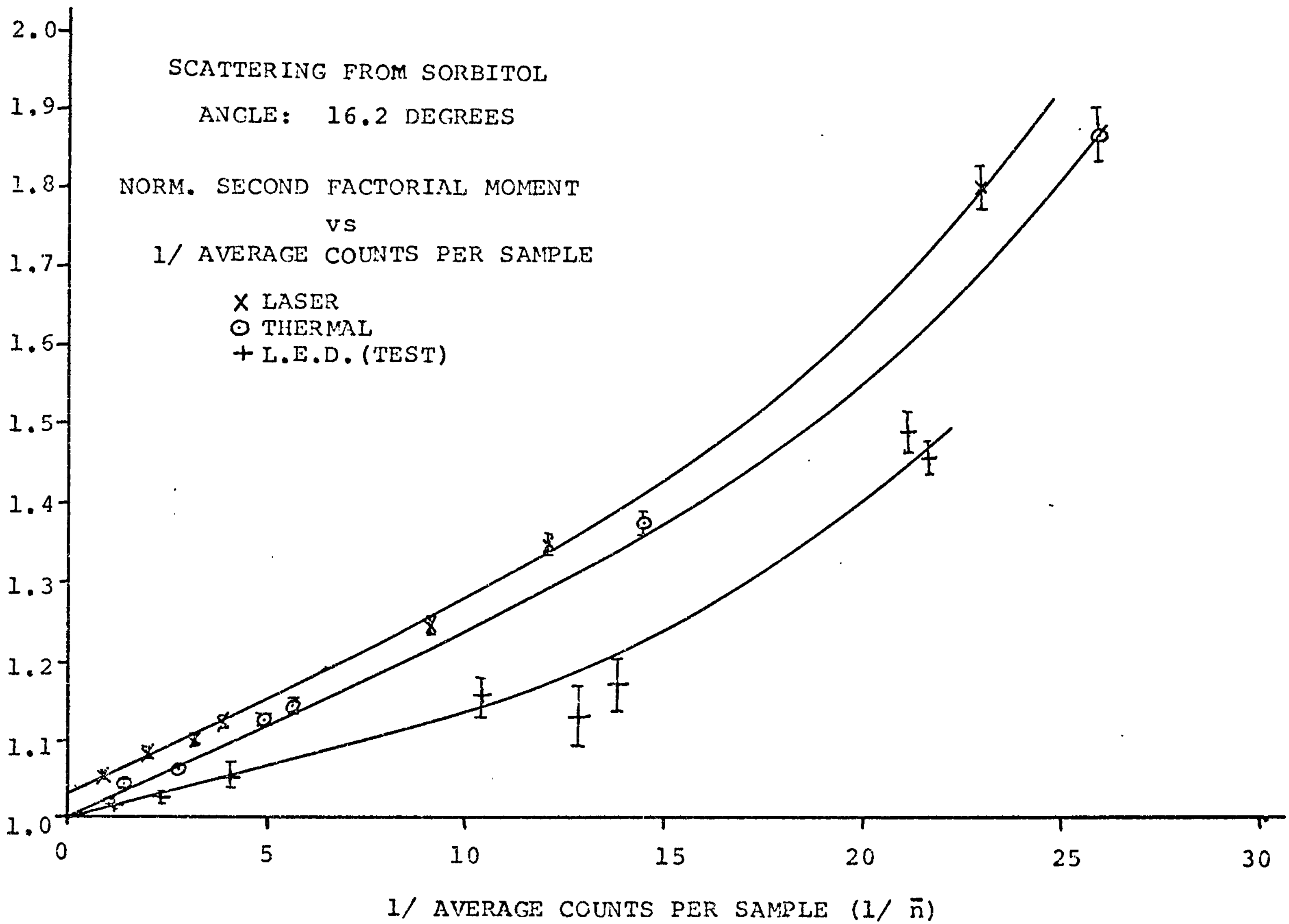

SCATTERING FROM SORBITOL
ANGLE: 16.2 DEGREES
NORM. SECOND FACTORIAL MOMENT
vs
1/ AVERAGE COUNTS PER SAMPLE
X LASER
⊙ THERMAL
+ L.E.D. (TEST)
2.0
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0
5
10
15
20
25
30
1/ AVERAGE COUNTS PER SAMPLE (1/ $\bar{n}$)

FIGURE 4

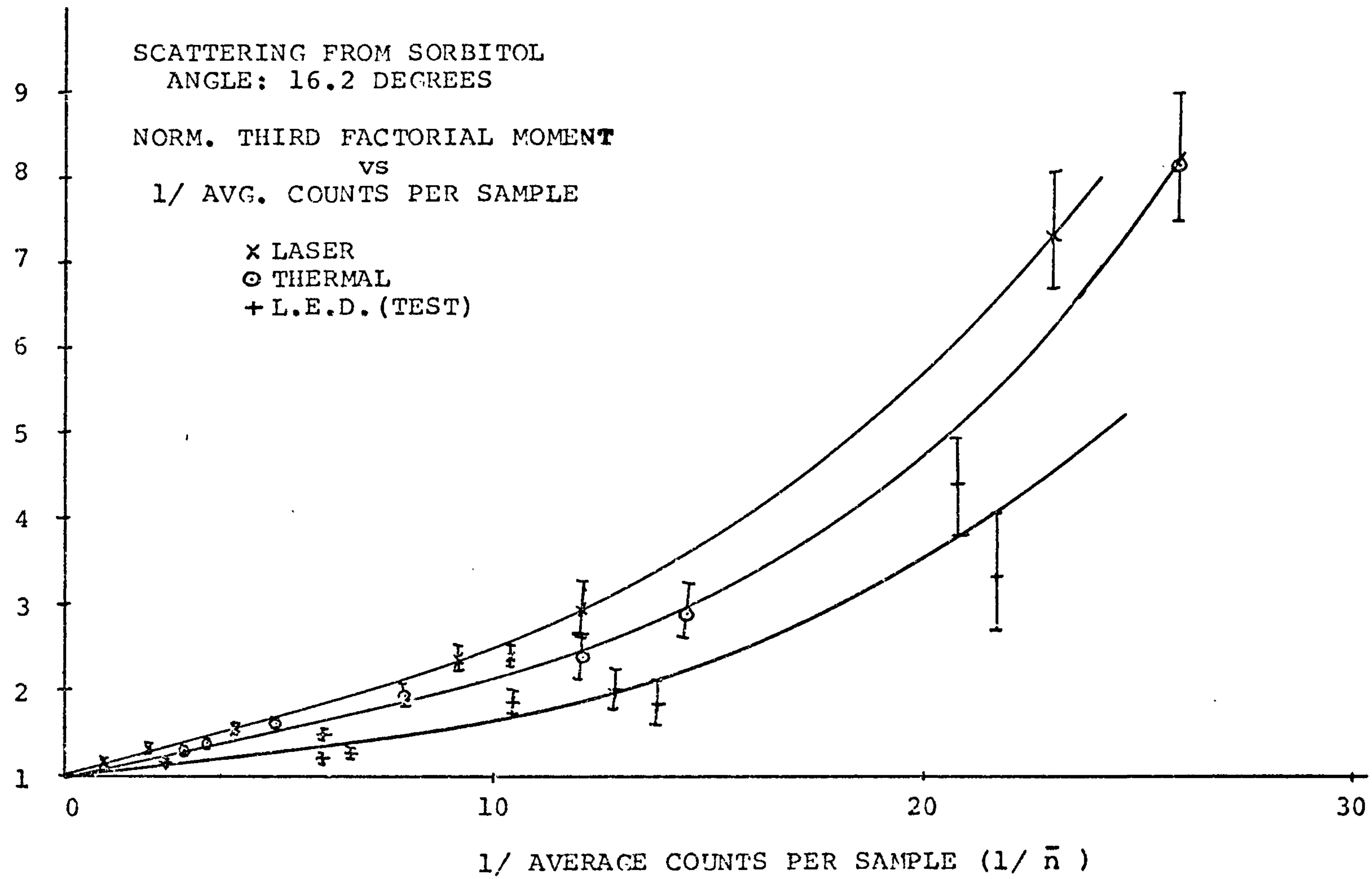
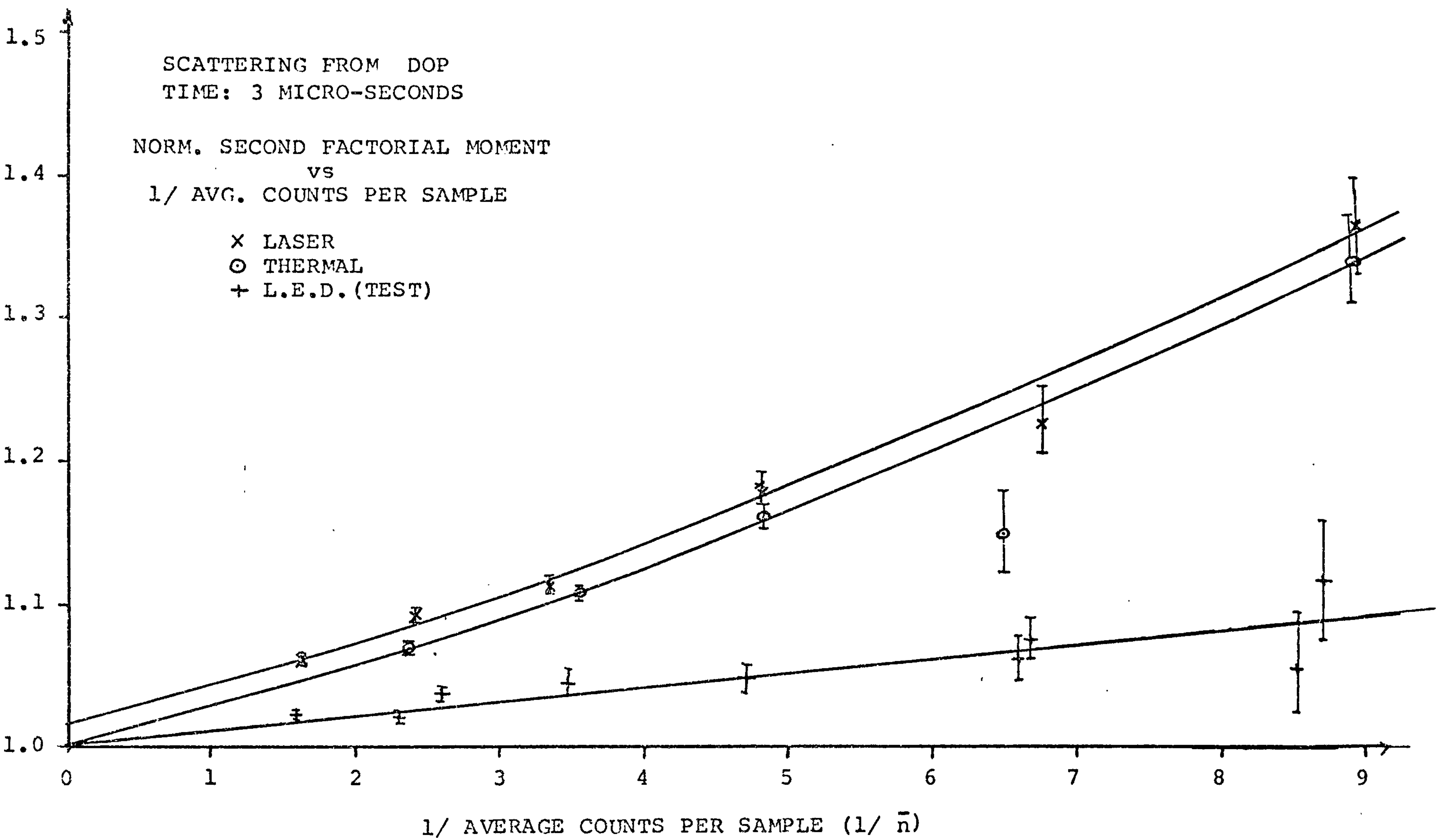
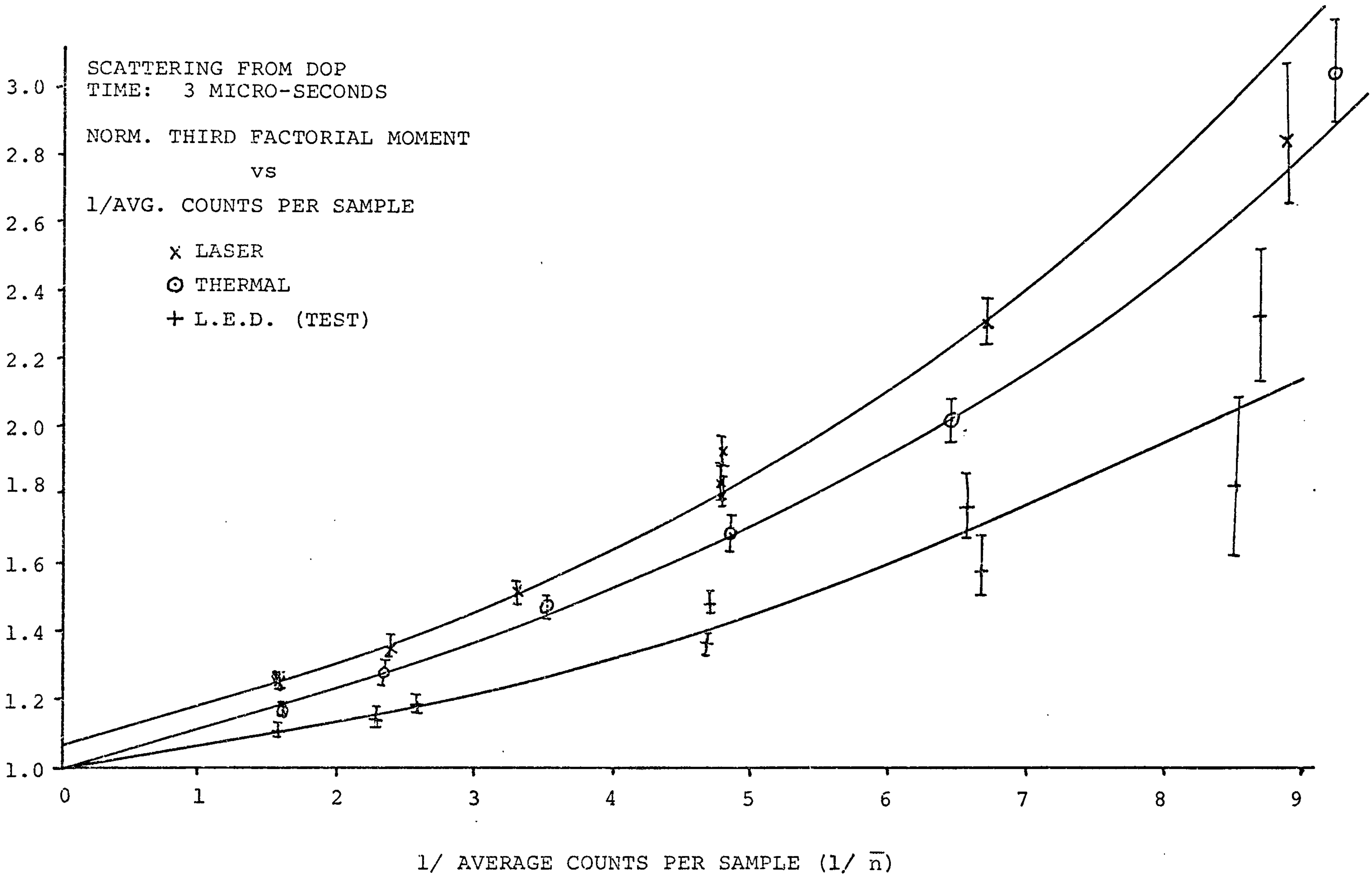

SCATTERING FROM SORBITOL
ANGLE: 16.2 DEGREES
NORM. THIRD FACTORIAL MOMENT
vs
1/ AVG. COUNTS PER SAMPLE
× LASER
⊙ THERMAL
+ L.E.D. (TEST)
1
2
3
4
5
6
7
8
9
0
10
20
30
1/ AVERAGE COUNTS PER SAMPLE (1/ $\bar{n}$)

FIGURE 5



SCATTERING FROM DOP
TIME: 3 MICRO-SECONDS
NORM. SECOND FACTORIAL MOMENT
vs
1/ AVG. COUNTS PER SAMPLE
× LASER
⊙ THERMAL
+ L.E.D. (TEST)
1.5
1.4
1.3
1.2
1.1
1.0
0
1
2
3
4
5
6
7
8
9
1/ AVERAGE COUNTS PER SAMPLE (1/ $\bar{n}$)

FIGURE 6



SCATTERING FROM DOP
TIME: 3 MICRO-SECONDS
NORM. THIRD FACTORIAL MOMENT
vs
1/AVG. COUNTS PER SAMPLE
× LASER
⊙ THERMAL
+ L.E.D. (TEST)
3.0
2.8
2.6
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0
1
2
3
4
5
6
7
8
9
1/ AVERAGE COUNTS PER SAMPLE (1/ $\overline{n}$)

FIGURE 7

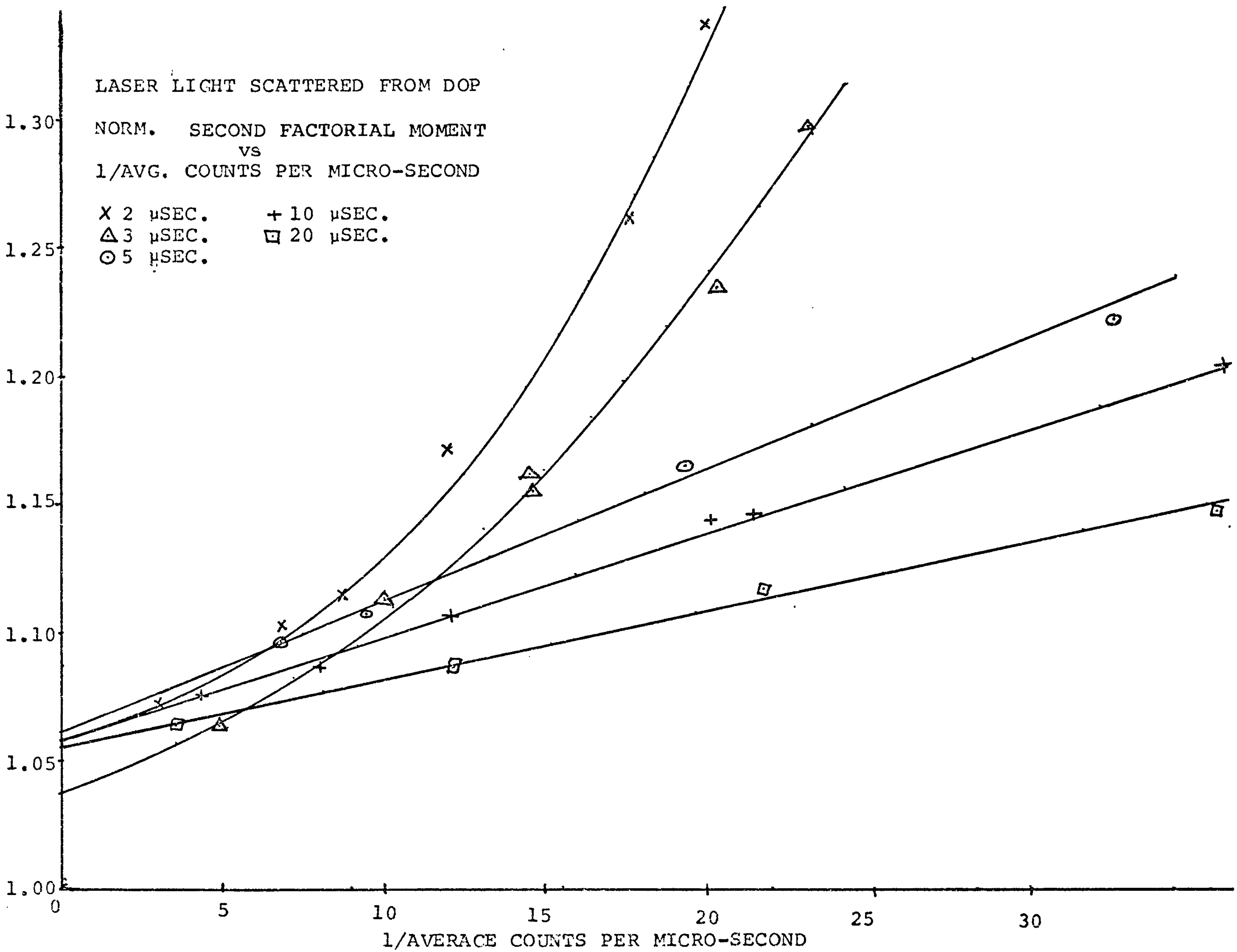
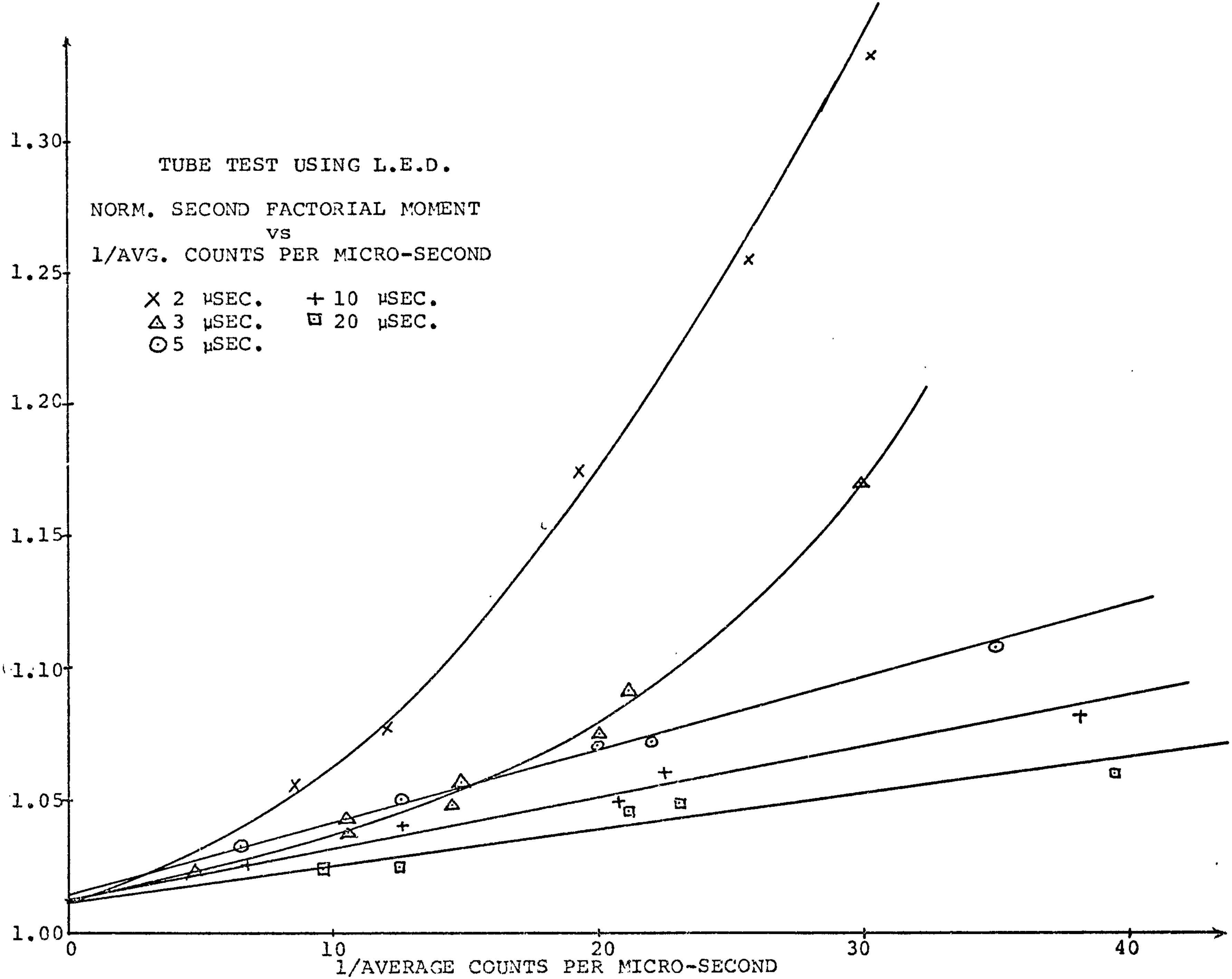

LASER LIGHT SCATTERED FROM DOP
NORM. SECOND FACTORIAL MOMENT
vs
1/AVG. COUNTS PER MICRO-SECOND
X 2 μSEC.
+ 10 μSEC.
Δ 3 μSEC.
□ 20 μSEC.
⊙ 5 μSEC.
1.30
1.25
1.20
1.15
1.10
1.05
1.00
0
5
10
15
20
25
30
1/AVERAGE COUNTS PER MICRO-SECOND

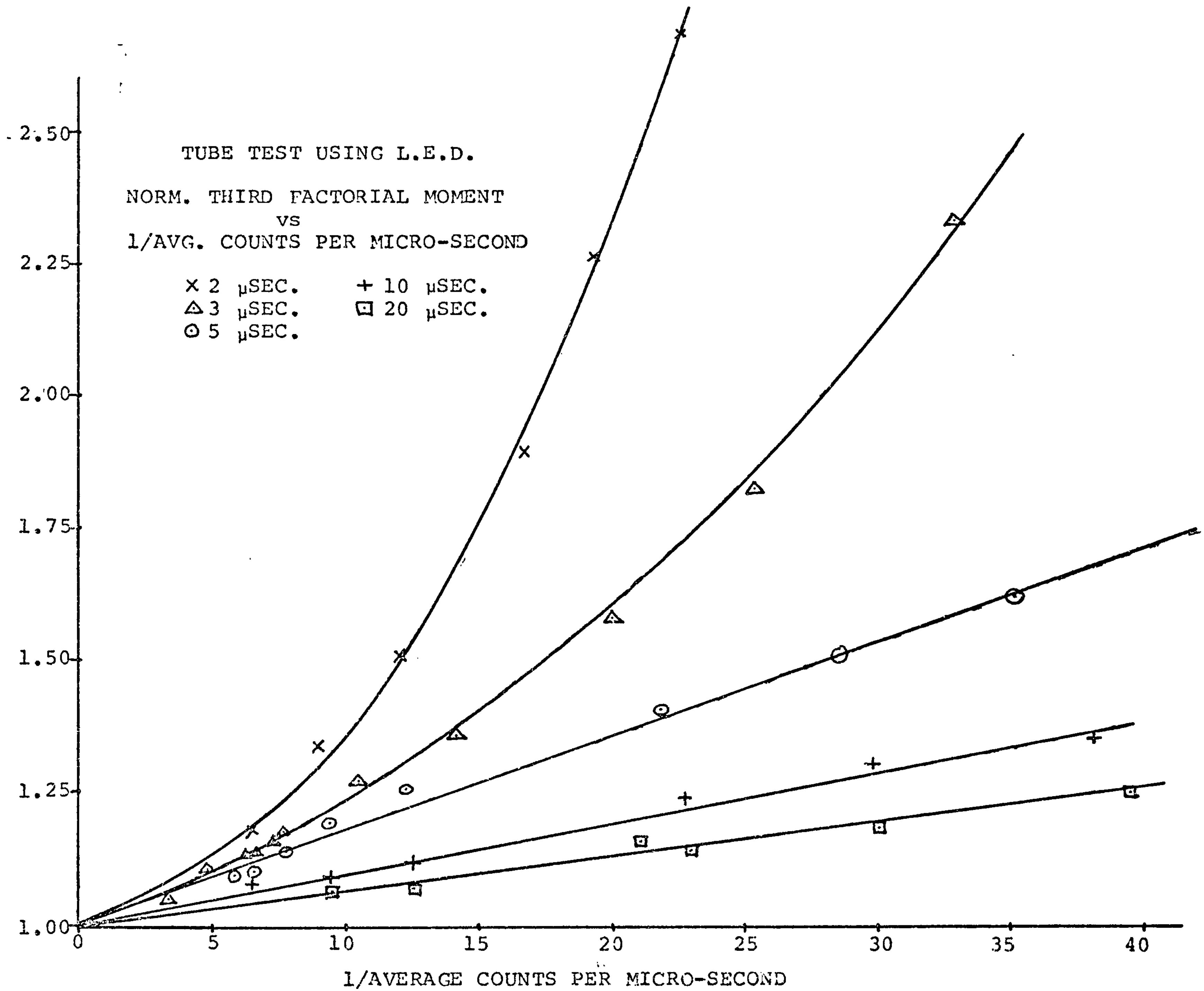
FIGURE 8



TUBE TEST USING L.E.D.
NORM. SECOND FACTORIAL MOMENT
vs
1/AVG. COUNTS PER MICRO-SECOND
× 2 μSEC.
△ 3 μSEC.
⊙ 5 μSEC.
+ 10 μSEC.
⊡ 20 μSEC.
1.30
1.25
1.20
1.15
1.10
1.05
1.00
0
10
20
30
40
1/AVERAGE COUNTS PER MICRO-SECOND

FIGURE 9

FIGURE 10



TUBE TEST USING L.E.D.
NORM. THIRD FACTORIAL MOMENT
vs
1/AVG. COUNTS PER MICRO-SECOND
× 2 μSEC.
△ 3 μSEC.
⊙ 5 μSEC.
+ 10 μSEC.
⊡ 20 μSEC.
2.50
2.25
2.00
1.75
1.50
1.25
1.00
0
5
10
15
20
25
30
35
40
1/AVERAGE COUNTS PER MICRO-SECOND

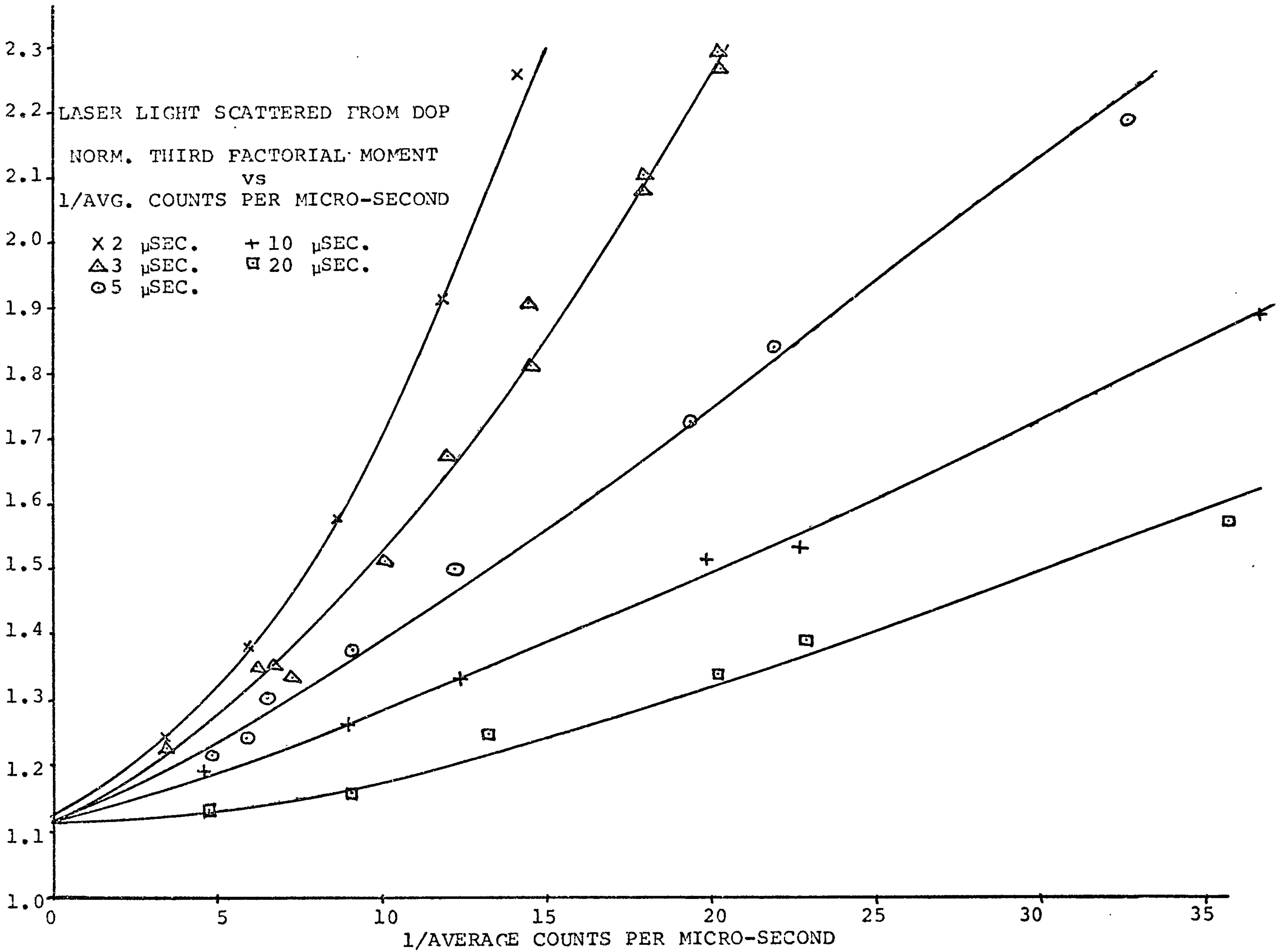

LASER LIGHT SCATTERED FROM DOP
NORM. THIRD FACTORIAL MOMENT
vs
1/AVG. COUNTS PER MICRO-SECOND
2 μSEC.
3 μSEC.
5 μSEC.
10 μSEC.
20 μSEC.
2.3
2.2
2.1
2.0
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0
5
10
15
20
25
30
35
1/AVERAGE COUNTS PER MICRO-SECOND

FIGURE 11

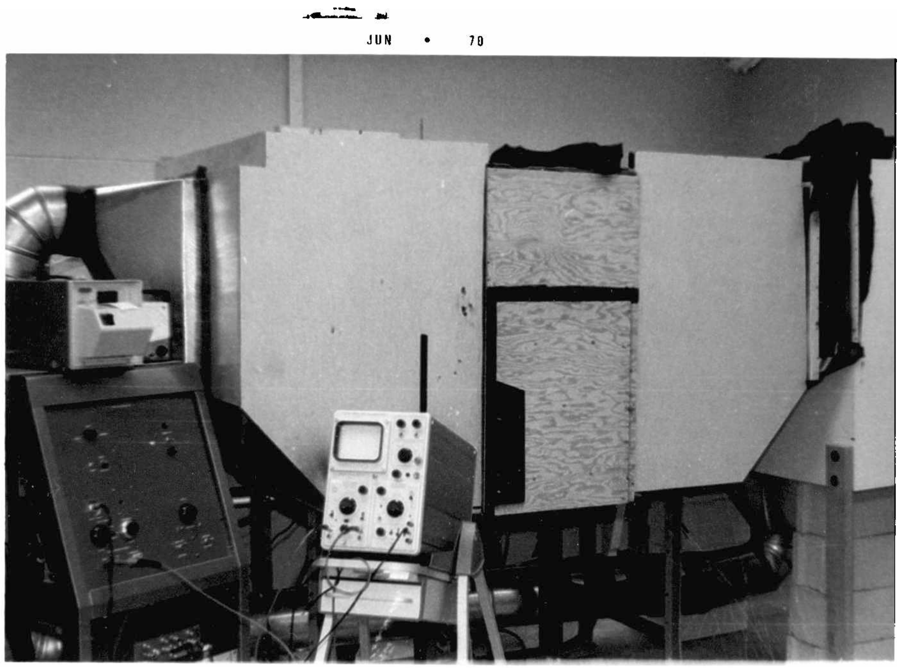
APPENDIX B

FORTRAN PROGRAM TO PROCESS DATA

```
      DIMENSION A(10),P(10),SU(10),SUN(10),VAR(5),STD(5)
     1SUNN(5),C(10)
      KODE = 0
      ANGLE = 0.0
      TIME = 0.0
  101 READ(5,1)KOD,AN,T,(A(I),I=1,10),SC1,SC2,SC3
    1 FORMAT(I1,2F5.1,2F6.0,2F5.0,2F4.0,2F3.0,2F2.0,3A4)
      IF(KOD.NE.KODE.OR.AN.NE.ANGLE.OR.T.NE.TIME) GO TO 102
      DO 103 I= 1,10
  103 C(I) = C(I) + A(I)
      GO TO 101
  102 IF (TIME .EQ. 0.0) GO TO 210
      SUM1 = 0.0
      SUM2 = 0.0
      DO 10 I = 1,10
      SUM1 = SUM1 + C(I)
   10 SUM2 = SUM2 + C(I)*C(I)
      AVG = SUM2/SUM1
      CPS = AVG * (10.**6)/ TIME
      NUM = SUM1
   15 DO 16 I = 1,10
   16 P(I) = C(I)/SUM1
   17 DO 20 L= 1,6
   18 SU(L) = 0.0
   19 DO 20 N = L,9
   20 SU(L) = SU(L) + FA(N)*P(N+1)/FA(N-L)
   21 DO 22 I = 1,4
   22 SUN(I) = SU(I)/(SU(1)**I)
   23 DO 30 J = 1,3
   24 VAR(J) = - SU(J)*SU(J)
   25 L = J+1
   26 DO 28 K = 1,L
   27 N = K-1
   28 VAR(J)=VAR(J)+((FA(J)/FA(N)/FA(J-N)**2)*SU(2*J-N)*FA(N)
   29 STD(J) = SQRT(VAR(J)/SUM1)
   30 SUNN(J) = STD(J)/(SU(1)**J)
      GO TO (201,202,203) KODE
  201 WRITE(6,111) ANGLE, TIME, PID1,PID2,PID3
      GO TO 204
  202 WRITE(6,112) ANGLE, TIME, PID1,PID2,PID3
      GO TO 204
  203 WRITE(6,113) TIME, PID1, PID2, PID3
  204 WRITE(6,110)NUM,CPS,SU(1),STD(1),SU(2),STD(2),SU(3),
     1 STD(3),SUN(2),SUNN(2),SUN(3),SUNN(3),KODE
```

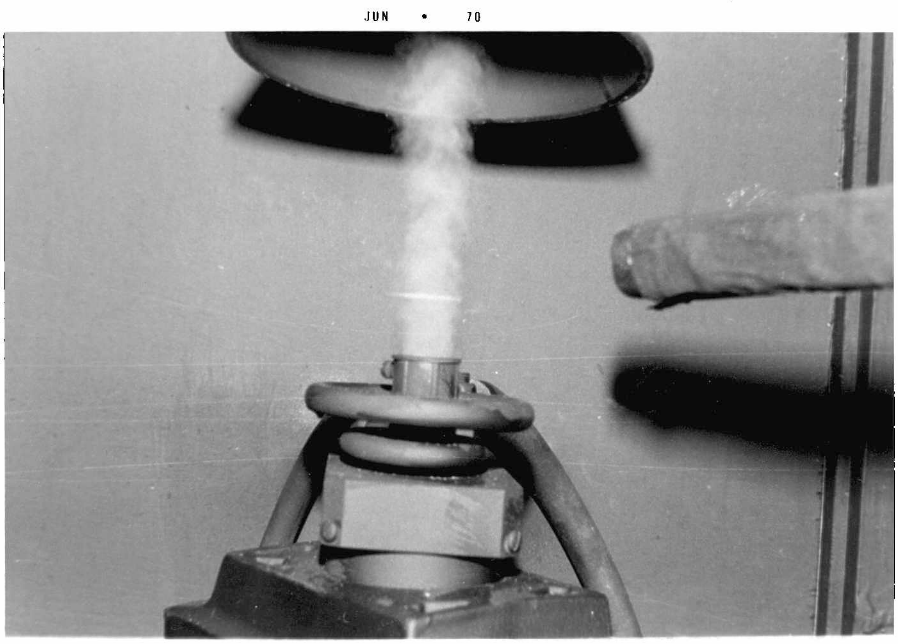
```
111 FORMAT(//10X,'LASER SCATTER ANGLE = ',F5.1,10X,'TIME = ',
   1 F5.1,5X,3A4/)
112 FORMAT(//10X,'THERMAL SCATTER  ANGLE = ',F5.1,10X,
   1 'TIME = ',F5.1,5X,3A4/)
113 FORMAT(//10X,'TUBE TEST',20X,'TIME = ',F5.1,3A4/)
110 FORMAT(2X,I10,2X,E10.4,3(1X,E10.4,'*',E9.4),2(1X,F8.4,
   1 '*',F8.4),2X, I5)
210 KODE = KOD
    TIME = T
    ANGLE = AN
    PID1 = SC1
    PID2 = SC2
    PID3 = SC3
    DO 216 I = 1,10
216 C(I) = A(I)
    GO TO 101
    STOP
    END


    FUNCTION FA(N)
    FA = 1.0
    IF (N .LE. 1) GO TO 95
    DO 94  J = 1,N
 94 FA = FA * J
 95 RETURN
    END
```

APPENDIX C



SCATTERING CHAMBER EXTERNAL VIEW

FIGURE 1



AEROSOL FLOW SYSTEM

FIGURE 2

AEROSOL GENERATOR

FIGURE 3

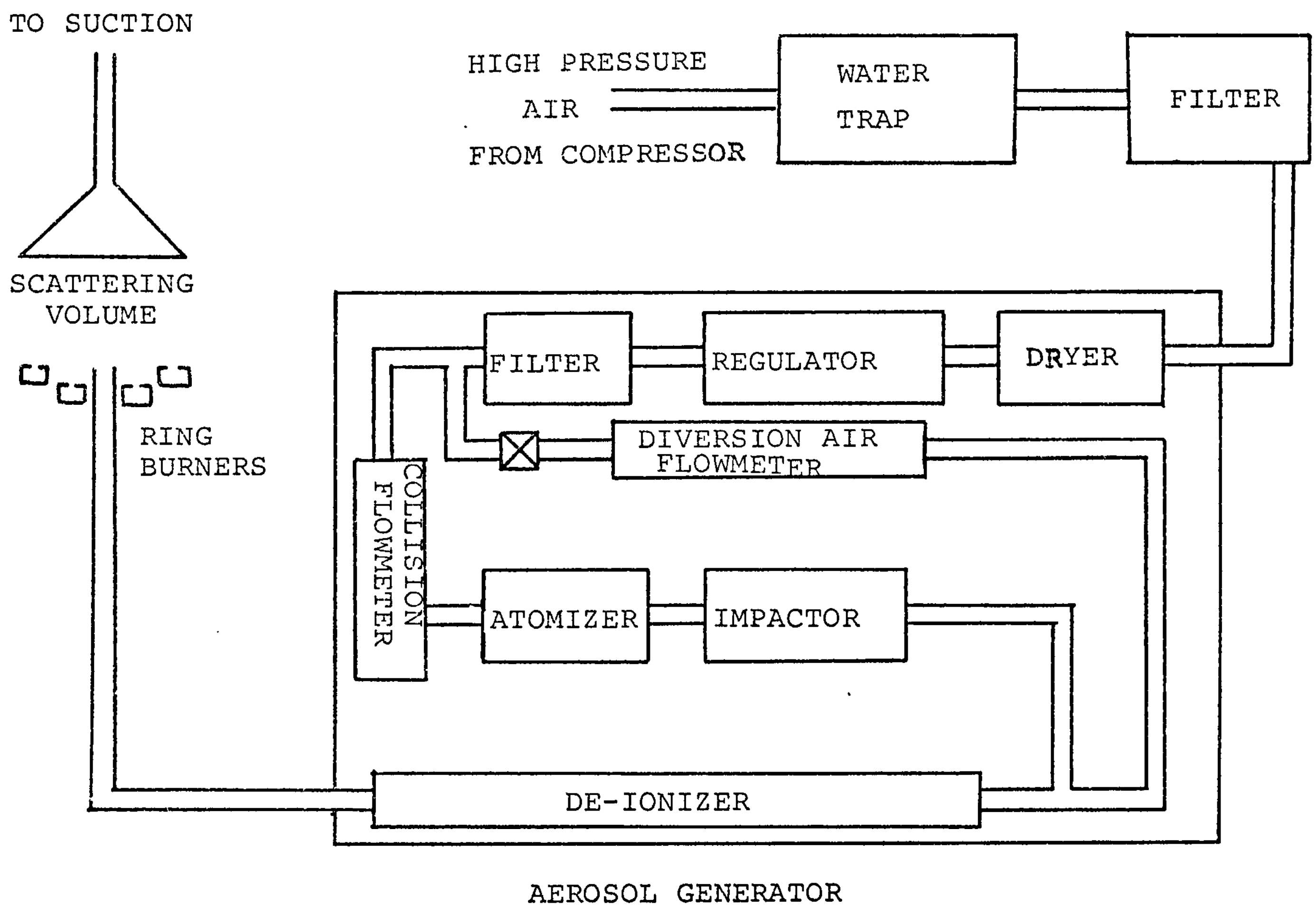
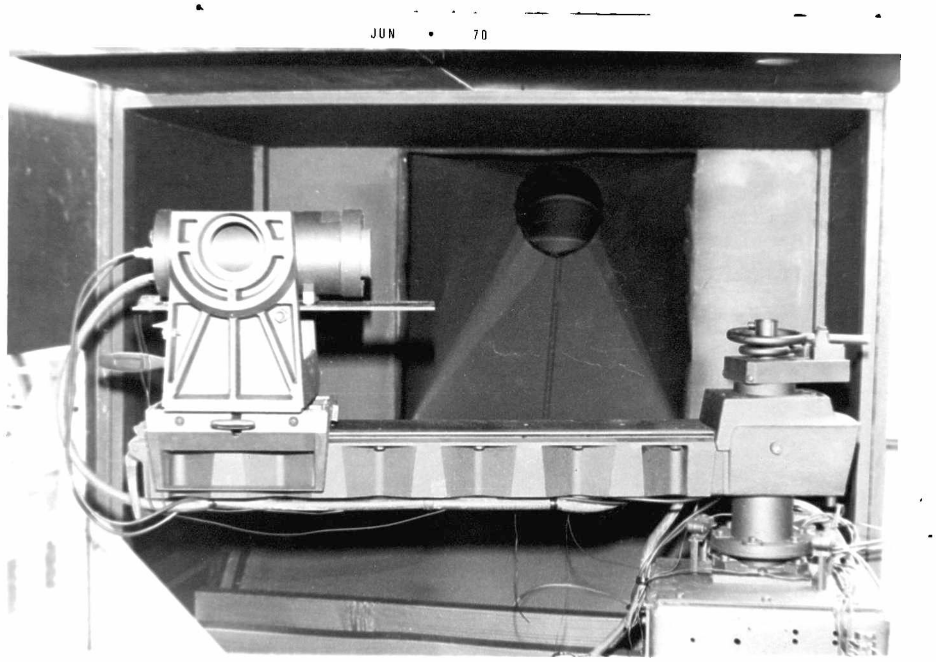


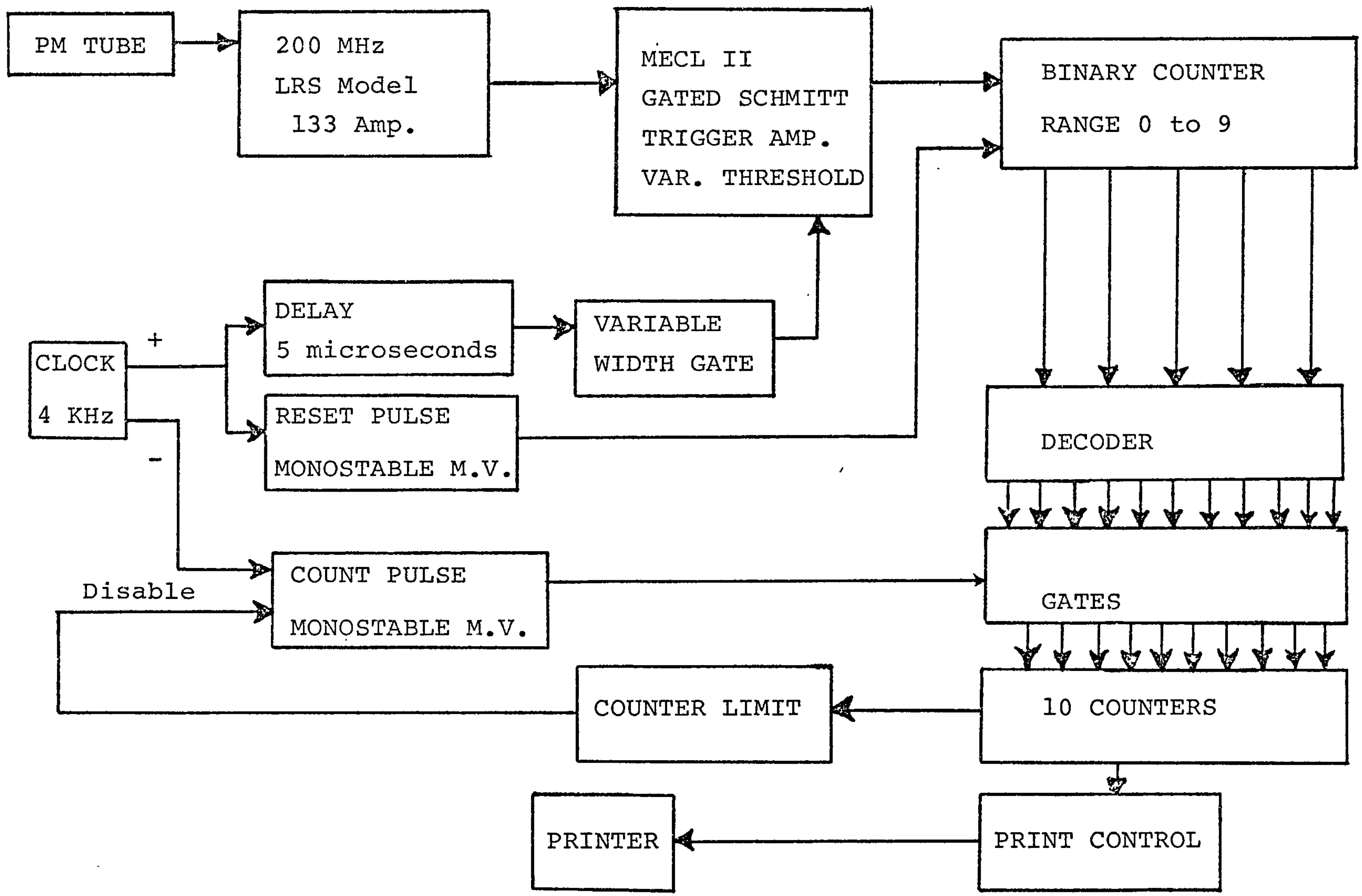
FIGURE 4

DIAGRAMMATIC REPRESENTATION OF THE AEROSOL FLOW SYSTEM



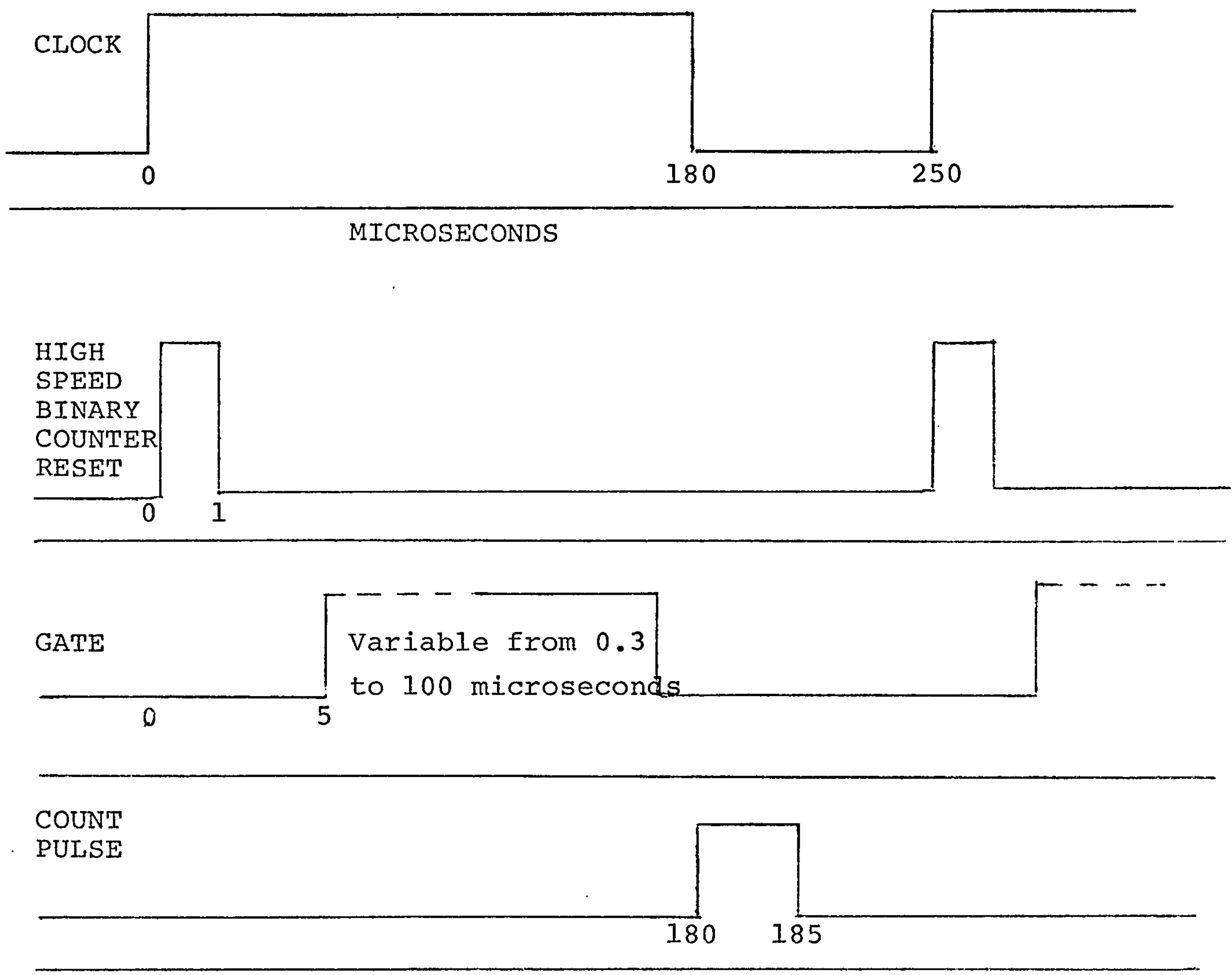
DETECTOR MOUNT

FIGURE 5



ELECTRONIC SYSTEM BLOCK DIAGRAM

FIGURE 6



SYSTEM TIMING DIAGRAM

FIGURE 7

Illustration of Printer Output Format

Illustration	Example
9 y γ δ	9 9 6 6
8 x α β	8 8 9 0
7 v w η	7 2 7 3
6 t u θ	6 3 6 2
5 q r s	5 2 4 5
4 m n p	4 9 5 4
3 j k l	3 2 7 6
2 g h i	2 4 5 7
1 d e f	1 3 8 3
0 a b c	0 1 8 1

Channel No.	Illustration	Example
0	a b c α β	1 8 1 9 0
1	d e f γ δ	3 8 3 6 1
2	g h i θ	4 5 7 2
3	j k l η	2 7 6 3
4	m n p	9 5 4
5	q r s	2 4 5
6	t u	3 6
7	v w	2 7
8	x	8
9	y	9

Figure 8

INSTRUMENT PANEL

FIGURE 9

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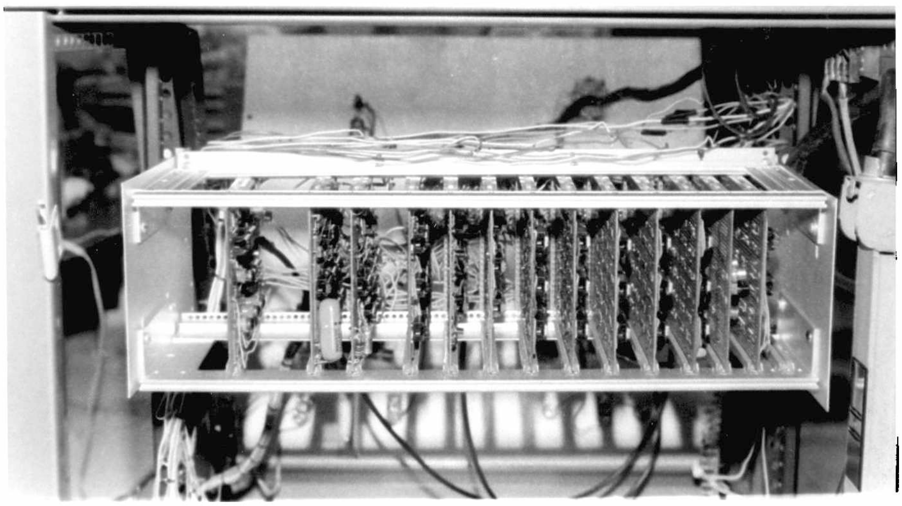

INTEGRATED CIRCUIT RACK

FIGURE 10

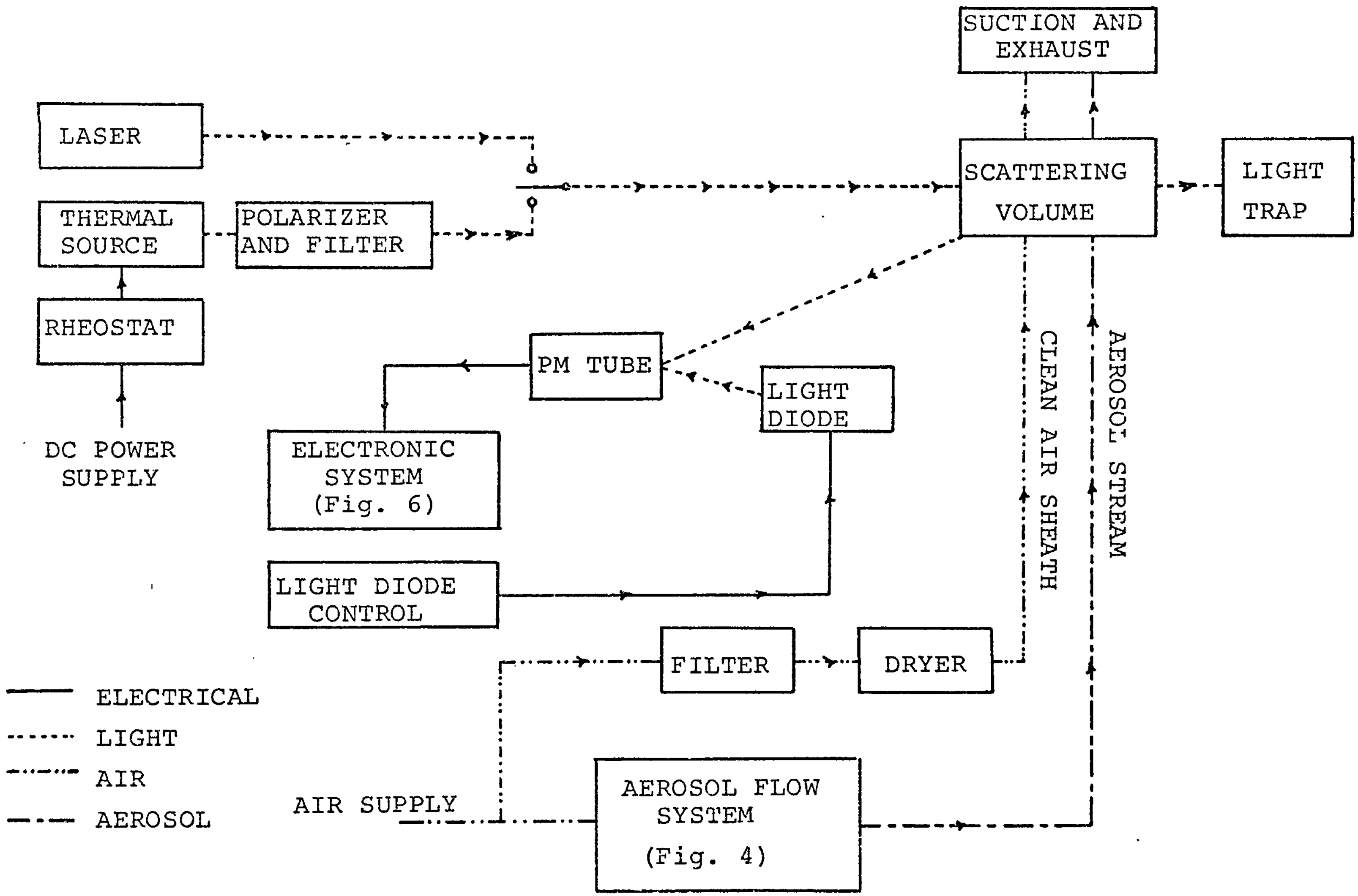

FIGURE 11. LINE DIAGRAM OF EXPERIMENTAL SET-UP