

by the radiator area given by its overall dimensions multiplied by  $\sigma T^4$ .

In face of these irregularities existing in a machined grooved surface, the error introduced by the first three assumptions will be small for all practical purposes. However, it might become significant at very small angles, i.e., smaller than  $10^\circ$ .<sup>5,6</sup>

It should be kept in mind that the depth of the grooves should be kept small so that the walls remain isothermal. To make the grooves too small, however, introduces new difficulties. The roundness at the top becomes significant; therefore, the surfaces of the grooves, on the average, are out of flat. Here again, experiments have indicated that, if  $(r/d) < 5\%$ , the relationships discussed hold well.

In conclusion, it can be stated that the application of V-shaped grooves on a metallic surface will increase its thermal emissivity in a practicable manner.

The assumptions made initially are justified for all practical purposes, considering the difficulties presented in the analysis when assumptions 1-3 are dropped.

It is important to point out that a groove of less than  $45^\circ$  angle provides apparent emissivities two to four times greater

than the surface emissivities. For base metals this is quite significant, since most of them exhibit total thermal emissivities of less than 0.2.

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# Measurement of Mean Particle Sizes of Sprays from Diffractively Scattered Light

R. A. DOBBINS,\* L. CROCCO,† AND I. GLASSMAN‡  
*Princeton University, Princeton, N. J.*

**The angular distribution of scattering for polydispersions of particles distributed according to the Upper Limit Distribution Function is examined and is found to lack the sensitivity necessary to permit determination of size distribution. However, the volume-to-surface mean diameter is found to be directly dependent upon angular distribution of intensity for a wide variety of shapes of the distribution function. Therefore, the combination of both a scattering experiment together with a transmission experiment can be used to obtain both particle concentration and volume-to-surface mean diameter of particles in a spray. While there is no limitation with regard to the maximum diameter, the actual upper size limit that is measurable experimentally is controlled by considerations related to angular resolving power. Experimental results that show agreement between the volume-to-surface mean diameter as determined by scattering experiment and by microscopic examination are given for solid spheres.**

**T**HE measurement of particle sizes present in liquid sprays poses a problem of considerable importance that has received frequent attention in the past. In the presence of evaporation of the liquid medium, only the use of high-speed photomicrography to examine the particles in situ is appropriate as a means of determining particle sizes.<sup>1,2</sup> The method has various difficulties related to the high time resolution required, e.g., the very small depth of focus of the

microscope, the small number of particles present within the focal plane, and the question of whether these particles constitute a representative sample. The careful application of the technique undoubtedly yields the most accurate results, and the method may be thought of as the primary standard of measurement in the present instance. On many occasions the awkwardness of application of the primary standard is an overwhelming drawback to its frequent use. It is, therefore, natural to inquire about other methods that might be used to determine the particle sizes present in sprays, methods that, although lacking the precision of the primary standard, do possess the advantages common to secondary methods.

A technique based on the scattering properties of the particles appears to fulfill the forementioned description, and investigation of such a technique first was performed by Chin, Sliepcevich, and Tribus<sup>3,4</sup> using a theory due to Gumprecht and Sliepcevich<sup>5</sup> which described the scattering properties of a polydispersion. This theory is applicable for polydispersions of such low concentration and/or limited spatial distribution so as to constitute a small optical depth and fur-

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\* Formerly Graduate Student, now Assistant Professor of Engineering, Brown University, Providence, R. I. Student Member AIAA.

† Robert H. Goddard Professor, Guggenheim Laboratories for the Aerospace Propulsion Sciences. Fellow Member AIAA.

‡ Associate Professor of Aeronautical Engineering, Guggenheim Laboratories for the Aerospace Propulsion Sciences. Member AIAA.

ther requires that both particle size and refractive index fall within given intervals. The formulation of a theory for the scattering properties in the more general case of particles of arbitrary large size and arbitrary refractive index occurring in polydispersions of finite optical depth has been discussed by the present authors.<sup>6</sup> The present paper reports the results of the application of this theory to the determination of particle sizes in a polydispersion.

The theory for the scattering of light by a single dielectric spherical particle of size number  $\alpha$  equal to  $\pi D/\lambda$ , where  $D$  is the diameter and  $\lambda$  is the wavelength of incident light, and refractive index  $m$  gives the following expression for the radiant intensity  $I(\theta)$  scattered at a small angle  $\theta$  measured from the forward direction due to an incident planar wave of irradiance  $E_0$ :

$$\frac{I(\theta)}{E_0} = \frac{D^2}{16} \left\{ \alpha^2 \left[ \frac{2J_1(\alpha\theta)}{\alpha\theta} \right]^2 + \left[ \frac{4m^2}{(m^2 - 1)(m + 1)} \right]^2 + 1 \right\} \quad (1)$$

where  $J_1$  is the Bessel function of first kind of order unity.

The three terms in the bracket of Eq. (1) represent, respectively, the Fraunhofer diffraction, the optical scattering due to refraction of the centrally transmitted ray, and the optical scattering due to a grazingly incident ray. Equation (1) requires that 1) the incident radiation is planar and monochromatic, 2) the forward angle  $\theta$  is small, 3) the particle size number  $\alpha$  and phase shift  $2\alpha(m - 1)$  are large, 4) the distance between particle and observer is large compared to  $D^2/\lambda$ , and 5) the particle is nonabsorbing.

If a polydispersion of particles is present, the integrated intensity of all particles is found by summing over all diameters. The relative frequency of occurrence of particles of a given diameter  $D$  are distributed according to a distribution function  $N_r(D)$  defined in such a way that the integral of  $N_r(D)$  over a given diameter interval represents the probability of occurrence of particles within the specified interval. When the expression for the intensity of scattering due to a polydispersion is normalized by dividing by the intensity of diffractively scattered light in the forward direction  $\theta = 0$ , then it is found that the second and third terms in Eq. (1) are small and can be ignored.<sup>6</sup> The normalized integrated intensity of forward scattered light  $I(\theta)$  due to a polydispersion of large particles therefore is given as

$$I(\theta) = \frac{\int_0^{D_\infty} \left[ \frac{2J_1(\alpha\theta)}{\alpha\theta} \right]^2 N_r(D) D^4 dD}{\int_0^{D_\infty} N_r(D) D^4 dD} \quad (2)$$

Equation (2) holds when all particles are illuminated equally or, in other words, when the attenuation of the incident beam is slight.

The transmission law for a polydispersion of large particles is

$$\frac{E}{E_0} = \exp \left\{ - \frac{\pi}{\pi} C_n l \int_0^{D_\infty} K(D, m) N_r(D) D^2 dD \right\} \quad (3)$$

The turbidity,  $\tau$ , is defined as

$$\tau \equiv \frac{\pi}{4} C_n \int_0^{D_\infty} K(D, m) N_r(D) D^2 dD \quad (4)$$

The mean scattering coefficient  $\bar{K}$  and the volume-to-surface mean diameter  $D_{32}$  also can be defined:

$$\bar{K} \equiv \frac{\int_0^{D_\infty} K(D, m) N_r(D) D^2 dD}{\int_0^{D_\infty} N_r(D) D^2 dD} \quad (5)$$

$$D_{32} \equiv \frac{\int_0^{D_\infty} N_r(D) D^3 dD}{\int_0^{D_\infty} N_r(D) D^2 dD} \quad (6)$$

The volume concentration  $C_v$  is related to the number concentration  $C_n$  by

$$C_v = \frac{\pi}{6} C_n \int_0^{D_\infty} N_r(D) D^3 dD \quad (7)$$

In view of Eqs. (4-7), the transmission law can be expressed as

$$E/E_0 = \exp(-\tau l) = \exp \left[ - \frac{3}{2} (\bar{K} C_n l / D_{32}) \right] \quad (8)$$

The restriction on Eq. (2) that the particles are illuminated equally is fulfilled when the optical depth  $\tau l$  is small compared to unity.

However, when the effects of finite optical depth are considered, it is found that the influence of multiple scattering on the illumination profile is relatively weak, even at moderate optical depths.<sup>6</sup> This result can be explained by noting that the amount of light diffractively scattered by a large particle is one-half of the total light scattered and that the diffractive scattering is confined to a small angle of magnitude  $\lambda/D$  and is, therefore, extremely intense. Attenuation of the primary scattering occurs equally at all small angles and angular distribution remains undistorted. Distortion of the illumination profile results when diffractively scattered light is attenuated to such an extent that it is no longer large compared to the intensity of the more nearly isotropic multiple scattering. Since the angle containing a fixed fraction of total diffractively scattered light is inversely proportional to particle size, then optical depth at which significant distortion of the illumination profile,  $I(\theta)$  vs  $\theta$ , occurs will be larger as the particle size increases. Measurements with  $\lambda = 0.546\mu$  have indicated that for a  $D_{32}$  of  $145\mu$  the distortion of the illumination profile is slight at an optical depth of 2.0, and serious distortion of  $I(\theta)$  for a polydispersion of  $D_{32}$  equal to  $23.9\mu$  resulted at an optical depth of 3.0. In general, it is recommended that the optical depth be maintained below 1.5 in order to assure absence of adverse distortion of the illumination profile.

Thus Eq. (2) represents a relationship between the angular distribution of scattered light in terms of particle size distribution. The question now is posed as to whether a knowledge of angular distribution of scattered light  $I(\theta)$ , as determined experimentally, can be used to infer something about particle size distribution. This question is resolved directly by examining the illumination profiles,  $I(\theta)$ , for various distribution functions representative of those of interest.

The suitability of various algebraic functions in describing experimentally measured droplet size distributions has been discussed in detail by Mugele and Evans,<sup>7</sup> who point out the inadequacy of the conventional exponential types of distribution functions, e.g., Rosin-Rammler or Nukiyama-Tanasawa expressions, that do not admit a maximum particle size. Mugele and Evans show that choosing the parameters in these distribution functions in such a manner as to fit a size histogram most closely can predict a completely erroneous volume fraction curve or give erroneous values of the mean diameters. These shortcomings are overcome by the Upper Limit Distribution Function (ULDF) proposed by Mugele and Evans, a function that possesses the property that no particles exist of sizes larger than a specified  $D_\infty$ . The superiority of the function in representing experimental data of droplet size distributions has been demonstrated.<sup>7</sup>

The ULDF written in a form consistent with the definition of  $N_r(D)$  is

$$N_r(D) = C \frac{\exp \left\{ - \delta \ln \left[ \frac{aD}{D_\infty - D} \right] \right\}^2}{D^4 (D_\infty - D)} \quad (9)$$

where  $C$  is defined such that

$$\int_0^{D_\infty} N_r(D) dD = 1$$

The parameters  $a$  and  $\delta$  of Eq. (9) are evaluated to give the best fit for the experimentally determined size histogram, a

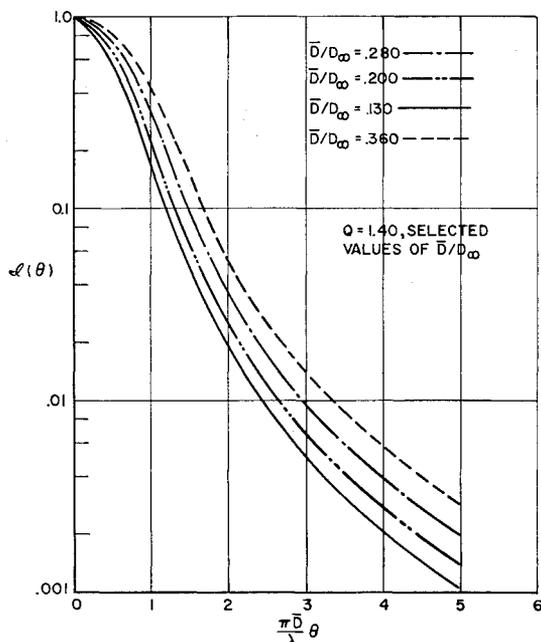


Fig. 1 Illumination profile for four size distributions of the ULDF type plotted against  $(\pi\bar{D}/\lambda)\theta$ .

process that is simplified by eliminating  $a$  and  $\delta$  in favor of two variables that possess a more direct geometric interpretation. Two such variables are the ratio of the most probable to maximum diameter,  $\bar{D}/D_\infty$ , and the half width or width of the distribution function at the two half peak values divided by the most probable diameter,  $Q$ . Designating the larger and smaller diameters that correspond to the two half peak values of the distribution function as  $D_{+1/2}$  and  $D_{-1/2}$ , respectively, the half width is defined as

$$Q \equiv (D_{+1/2} - D_{-1/2})/\bar{D} \quad (10)$$

Evaluating  $\bar{D}/D_\infty$  from Eq. (9) yields the following transcendental relation  $\bar{D}/D_\infty$  as a function of  $a$  and  $\delta$ :

$$a = \left(\frac{D_\infty}{\bar{D}} - 1\right) \exp\left[\frac{1}{\delta^2} \left(\frac{5}{2} \frac{\bar{D}}{D_\infty} - 2\right)\right] \quad (11)$$

The values of  $D_{+1/2}/\bar{D}$  and  $D_{-1/2}/\bar{D}$  are found from the following transcendental equation:

$$\left(\frac{D_{\pm 1/2}}{\bar{D}}\right)^4 \frac{D_\infty - D_{\pm 1/2}}{D_\infty - \bar{D}} = 2 \exp - \left[ \left(\delta \ln_e \frac{aD_{\pm 1/2}}{D_\infty - D_{\pm 1/2}}\right)^2 - \left(\delta \ln_e \frac{a\bar{D}}{D_\infty - \bar{D}}\right)^2 \right] \quad (12)$$

Examination of Eqs. (10-12) reveals that  $Q$  is a function only of  $a$  and  $\delta$ . Therefore, both shape parameters  $\bar{D}/D_\infty$  and  $Q$  are determined by specifying  $a$  and  $\delta$  and conversely.<sup>§</sup>

The various mean diameters,  $D_{PQ}$ , are defined in a general manner by the following

$$D_{PQ}^{P-Q} \int_0^{D_\infty} N_r(D) D^Q dD = \int_0^{D_\infty} N_r(D) D^P \quad (13)$$

Mugele and Evans have evaluated certain mean diameters; their results for the volume-to-surface mean diameter is

$$D_{32}/\bar{D} = (D_\infty/\bar{D}) [1 + a \exp(-1/4\delta)] \quad (14)$$

Thus it is found that  $D_{32}/\bar{D}$  is determined by specifying the shape parameters  $a$  and  $\delta$  or  $Q$  and  $\bar{D}/D_\infty$ . A similar result holds for all other mean diameters.

<sup>§</sup> Appropriate graphs for relating typical values of  $a$  and  $\delta$  to  $\bar{D}/D_\infty$  and  $Q$  have been formulated. They are not reproduced here for they should be examined in the context of Ref. 8, which will be submitted for publication as a Technical Note.

Substituting Eq. (9) into Eq. (2) gives the following relation for the forward scattering due to distribution of particle sizes obeying the ULDF:

$$I(\theta) = \frac{\int_0^{\zeta_\infty} \left[ \frac{2J_1(\alpha_0 \zeta)}{\alpha_0 \zeta} \right]^2 \frac{\zeta_\infty}{\zeta_\infty - \zeta} \exp - \left( \delta \ln \frac{a\zeta}{\zeta_\infty - \zeta} \right)^2 d\zeta}{\int_0^{\zeta_\infty} \frac{\zeta_\infty}{\zeta_\infty - \zeta} \exp - \left( \delta \ln \frac{a\zeta}{\zeta_\infty - \zeta} \right)^2 d\zeta} \quad (15)$$

where  $\zeta = D/D_0$ , and  $\alpha_0 = (\pi D_0/\lambda)\theta$ , and  $D_0$  is any convenient dimension, e.g.,  $\bar{D}$  or any  $D_{PQ}$ , used to form a non-dimensional diameter.

Equation (15) shows that, if  $D_0$  is chosen equal to  $\bar{D}$ , the illumination profile as a function of the reduced angle  $(\pi\bar{D}/\lambda)\theta$  depends only on the shape parameters. Similitude considerations therefore permit the investigation of a wide range of illumination profiles by a limited number of calculations for the relevant shape parameters. Illumination profiles have been calculated for 18 points scattered throughout the  $D/D_\infty$  (0.13-0.28)- $Q$  (0.50-2.10) plane in the range where the shape parameters describe positively skewed functions.

Shown in Fig. 1 is a number of illumination profiles for various values of  $\bar{D}/D_\infty$  with  $Q$  equal to 1.40. Examination of Fig. 1 shows that all of the theoretical illumination profiles possess the same general monotonic shape and lack any distinctive features that would permit recognition of different values of  $\bar{D}/D_\infty$  even if it is known, a priori, that  $Q$  is a known or unknown constant. Furthermore, Fig. 1 shows that curves appear affinely related and such a relationship is indeed valid to within a reasonable expectation of experimental accuracy. Since the abscissa contains the most probable diameter  $\bar{D}$  in the reduced angle, one is obliged to conclude that no unique combination of  $\bar{D}/D_\infty$  and  $\pi\bar{D}/\lambda$  can be inferred from a given measurement of  $I(\theta)$  vs  $\theta$ . This result was found to be true for various other values of  $Q$ , and, therefore, it is concluded that the scattered light profile lacks the sensitivity necessary to determine the shape parameters uniquely.

In view of the lack of uniqueness in determining the distribution function, an attempt was made to determine if the mean diameters  $D_{PQ}$  could be determined from a given profile illumination profile. This was done by successively allowing  $D_0$  to equal  $D_{32}$ ,  $D_{30}$ ,  $D_{21}$ , and  $D_{10}$ . In general, it was found that the illumination profiles were brought into near

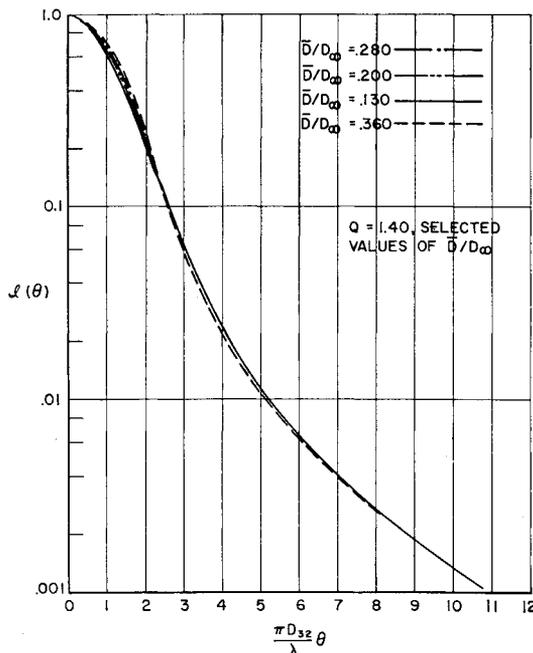
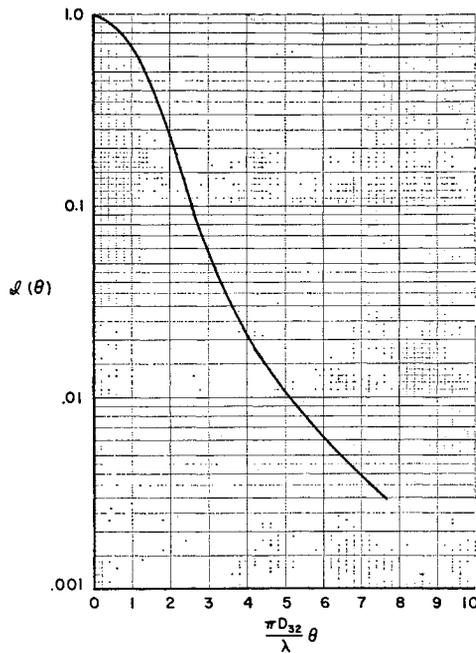


Fig. 2 Illumination profile for four size distributions of the ULDF type plotted against  $(\pi D_{32}/\lambda)\theta$ .



**Fig. 3 Mean theoretical illumination profile for polydispersions with sizes distributed according to the ULDF.**

coincidence with the best results obtained using  $D_{32}$ . Figure 2 shows the same profiles given in Fig. 1 but plotted against reduced angle  $(\pi D_{32}/\lambda)\theta$  and shows the profiles to be nearly coincident. A mean illumination profile for eighteen points in the  $Q-\bar{D}/D_\infty$  plane has been determined and is shown in Fig. 3 and tabulated in Table 1, where the standard deviation as a percentage of the mean value of  $I(\theta)$  for given value of angle is tabulated also. The variation of  $I(\theta)$  for given angle is considered very small in view of the logarithmic variation of this quantity. Furthermore, if one calculates the variation from the mean of the angle corresponding to a given value of  $I(\theta)$ , then the standard deviation will be reduced considerably below the values listed in Table 1. Thus the experimental determination of  $I(\theta)$  vs  $\theta$  does permit an evaluation of  $D_{32}$  if the distribution function is known to be an approximation to the ULDF.

Examination of Eq. (2) describing the integrated light scattering profile indicates that the relative contribution due to the various particle sizes is in proportion to  $D^4$ , and one concludes that the influence of the smaller particles in the polydispersion is negligible. This is substantiated by calculations of  $I(\theta)$  for two distribution functions of radically different shape to the left of the most probable diameter but otherwise identical. The resulting illumination profiles are so close as to be undistinguishable to an accuracy equal to that obtainable from experiment. Thus it is only necessary that the right-hand limb of the distribution function be an approximation to the ULDF in order that  $I(\theta)$  depend solely on  $D_{32}$ . This restriction is quite mild and one concludes that the dependency of  $I(\theta)$  on  $D_{32}$  must be more general than to apply merely to polydispersions obeying the ULDF. The failure of the illumination profile to provide adequate information to describe the distribution functions of the polydispersion is disappointing. However, the conclusion that  $D_{32}$  can be determined by a scattering experiment is particularly fortunate because the knowledge of volume-to-surface mean diameter can be used together with results of an optical transmission test [Eq. (8)] to yield particle concentration.

A case of special interest arises when the distribution function is a Dirac function, i.e., when all particles are the same size. The expression for the illumination profile then reduces to

$$I(\theta) = [2J_1(\alpha\theta)/\alpha\theta]^2 \quad (16)$$

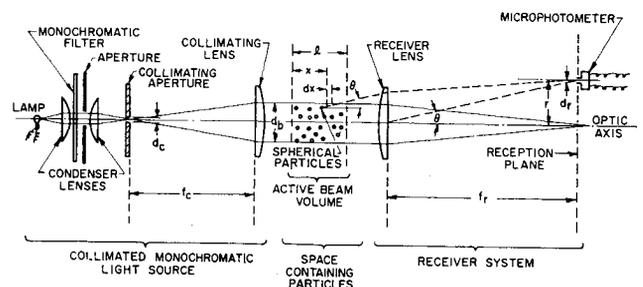
**Table 1 Mean theoretical illumination profile for polydispersions with sizes distributed according to the ULDF**

Reduced angle, $(\pi D_{32}/\lambda)\theta$	Mean <sup>a</sup> normalized illumination, $I(\theta)$	Standard deviation of mean $I(\theta)$ as a percentage of the mean value <sup>a</sup>
0	1.000	0
1.00	$6.86 \times 10^{-1}$	4.68
2.00	$2.33 \times 10^{-1}$	9.69
3.00	$5.56 \times 10^{-2}$	7.93
4.00	$2.06 \times 10^{-2}$	19.1
5.00	$1.06 \times 10^{-2}$	10.1
6.00	$6.05 \times 10^{-3}$	8.86
7.00	$3.74 \times 10^{-3}$	9.60
8.00	$2.48 \times 10^{-3}$	8.71

<sup>a</sup> Mean value is based on average of the value of  $I(\theta)$  for 18 ULDF with  $Q$  ranging from 0.5 to 2.1 and  $\bar{D}/D_\infty$  ranging from 0.13 to 0.28.

This equation describes the familiar ringed diffraction pattern, which permits a direct determination of particle size by measurement of the angle subtended by any one of the dark rings corresponding to the zeros of  $J_1(\alpha\theta)$  occurring at  $\alpha\theta = 3.83, 7.02, 10.17$ , etc. Schmidt<sup>9</sup> used this method of measuring particle sizes produced by a special type of nozzle but expressed concern as to the significance of the diameter measured if it was not known a priori that the particle size was uniform. This question has been resolved in the course of the present studies of theoretical illumination profiles due to polydispersions with size distribution described by the ULDF. It was found that the theoretical illumination profiles start to depart from the average profile in a manner that indicates the growth of a ringed diffraction pattern for values of  $Q$  less than one half. In this region of the  $Q - \bar{D}/D_\infty$  plane, all mean diameters,  $D_{PQ}/\bar{D}$  approach the value of unity. Therefore, the appearance of a ringed structure to the diffraction pattern indicates that the distribution function is sufficiently narrow so that all mean diameters are coincident with the most probable diameter.

Experiments have been conducted in order to test the forementioned results using the apparatus shown in Fig. 4. The focal lengths of the collimating and receiver lenses were 1000 and 1200 mm, respectively. A high-pressure mercury lamp of very high brightness was used as a lamp in order to produce a highly collimated beam of high irradiance. The microphotometer used a photomultiplier tube as a transducer. The diameter of the collimating aperture (0.79 mm) and the diameter of the microphotometer aperture ( $75\mu$ ) were chosen in order to afford the angular resolution necessary to measure the angular distribution of scattered light. That is, the diameter of the collimating aperture must be such that the divergence angle,  $\psi = d_c/2f_c$ , of the incident beam must be small compared to the angle of scattering. Whereas there is no limitation with regard to the maximum diameter in order that the forementioned theory be applicable, the actual upper size limit that is measurable is, in fact, controlled by considerations related to angular resolving power. As particle size increases, the angle containing a fixed fraction of the total diffractively scattered light decreases, and it becomes neces-



**Fig. 4 Schematic drawing of optical apparatus used to perform small angle scattering tests.**

**Table 2 Comparison of particle size measurement by observation of angular distribution of scattering with results of microscopic examination**

Particle group A, $D_{32} = 23.9\mu^a$		Particle group B, $D_{32} = 145\mu^a$	
$I(\theta)$	$D_{22},^b \mu$	$I(\theta)$	$D_{22},^b \mu$
0.53	26.6	0.60	142
0.45	25.2	0.52	137
0.30	25.0	0.405	146
0.195	24.7	0.35	147
0.130	23.9	0.21	143
0.079	24.4	0.126	146
		0.072	145

<sup>a</sup> By microscopic examination.

<sup>b</sup> Calculated from Fig. 3 using shown experimental value of  $I(\theta)$  from a single test.

sary to reduce the collimating aperture and therefore the amount of light irradiating the particles. The aperture in the microphotometer also must be reduced when the angle over which the scattering occurs is made smaller, and, ultimately, the ratio of the lamp brightness to transducer sensitivity determines the upper size limit that can be measured. Other important properties of this optical system with regard to its ability to integrate the light scattered by each particle and apparently to stop the motion of the particles have been described previously.<sup>6</sup>

Tests were conducted using glass and polystyrene spheres held in suspension in a liquid medium by a mild stirring action. The particles were taken from large batches whose particle size distribution had been determined in advance by preparing photomicrographs, measuring about 1000 particles of each batch, and preparing size distribution histograms from which the volume-to-surface mean diameter could be calculated. The results of measurements of  $D_{32}$  by scattering experiments are compared in Table 2 with the results of microscopic examination of the particles and show satisfactory agreement.

Additional experiments were performed using molten paraffin sprayed from a solid cone atomizing nozzle. Angular distribution of scattering was measured by a technique of photographic photometry described previously.<sup>10</sup> Three optical measurements were conducted at each of three atomizer pressures and the results are given in Table 3. Particle sizes also were measured by using glass slides to capture a sample of the particles which then were examined by photomicrographic techniques to determine size distribution. The data obtained by microscopic count unfortunately are not self-consistent in that no pressure effect is noted between the particles produced at 100 and 150 psi. The light scattering measurements do show a consistent trend toward a smaller particle size with higher atomizer pressure drop. In view of the self consistency of data obtained by light scattering measurements, the good agreement of the light scattering measurements on solid particles, and the inconsistent

**Table 3 Comparison of mean size of solidified sprayed particles of paraffin as determined by light scattering measurement and microphotographic count**

Atomizer pressure, psig	$D_{32}$ light scattering, $\mu$	$D_{32}$ by microphotography, $\mu$		
75	106	98	110	82
100	89	92	86	72
150	75	76	78	72

data obtained when the liquid particles were sampled on glass slides, it is concluded that the light scattering measurements gave the most accurate measurements of particle size. The error in the photomicrographic count is attributed to difficulties in obtaining a statistically good sample by capturing particles on a slide. A more extensive study of the atomization of liquids by an atomizing nozzle using the light scattering method just described has been performed by Webb and Cohen.<sup>11</sup>

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