THE USE OF LIGHT SCATTERING
FOR THE DETECTION OF FILTER SAMPLES OF FIBROUS AEROSOLS

Paul A. Baron, Ph.D.

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Center for Disease Control
National Institute for Occupational Safety and Health
Division of Physical Sciences and Engineering
Cincinnati, Ohio 45226

January 1978

DISCLAIMER

DHEW (NIOSH) Publication No. 78-105

ABSTRACT

The use of light scattering from fibrous dusts has been investigated. It has been found that straight fibers have a sufficiently distinctive scattering pattern that may be counted electronically. Curved fibers may also be detected by focussing the light source so as to illuminate only a small, relatively straight, section of the fiber. Theoretical and experimental information, as well as a proposed instrument, are presented in the report.

The proposed instrument would fill a need in industrial hygiene to obtain rapid, accurate measurements of personal exposures to asbestos and other fibrous dusts.

CONTENTS

INTRODUCTION

The accurate and precise analysis of industrial hygiene samples of fibrous aerosols, in particular asbestos, is difficult. The USPHS/NIOSH phase contrast light microscopy method[1] for counting asbestos fibers has been used in an inter-laboratory comparison to estimate the accuracy and precision of the method. It has been found that the precision for laboratory prepared samples is indicated by a coefficient of variation (CV) ranging from 0.30 to 1.0.[2] The accuracy cannot be estimated since there is currently no way of preparing known samples or of comparing this method directly with any other method. Because the toxicity of asbestos, which causes asbestosis and various types of cancer,[1] is so high, it is necessary to be able to detect and quantify extremely small amounts of material. The present Federal standard for asbestos specifies an environmental limit value of 2 fibers/cm^3.[3]

A reduced standard of 0.5 fibers/cm^3 has been proposed by the Occupational Safety and Health Administration (OSHA). When using accepted industrial hygiene personal sampling techniques, an asbestos aerosol concentration at the Federal standard can result in a sample mass in the range of 8 - 160 μg (see Appendix I). These sample levels have yet to be detected with demonstrated accuracy in industrial air samples by current analytical methods such as differential thermal analysis (DTA)[5] and X-ray diffraction (XRD).[6] While a detection limit (not defined) of about 10 μg has been claimed using a research DTA instrument, a detection limit (defined as one standard deviation) of 100 μg can be more reasonably be expected using a commercially available DTA instrument. The use of XRD for the detection of asbestos looks more promising. A recent study indicates a detection limit (defined as three times the noise level) of 10 μg by XRD for laboratory samples of asbestos mixed with alumina background. If the standard is reduced to 0.5 fibers/cm^3, even these state of the art detection limits may not be adequate. Apart from problems with sensitivity, DTA and XRD do not differentiate between species of asbestos and their non-fibrous polymorphs.

An additional problem is the relationship between the standard, which is in terms of a count of the fibers, and the results of TA and XRD, which are in terms of sample mass. The standard is based on toxicological and epidemiological data that relied on visual counting of asbestos fibers. The relationship between fiber count and total fiber mass cannot be accurately determined because of the highly variable size distribution of asbestos aerosols. This means that because of the problems of inadequate sensitivity and the lack of a fixed count to mass ratio for asbestos, it is unlikely that the standard will be modified to include a mass standard in the near future.

The phase contrast light microscopy method for counting asbestos fibers has been written into the Federal standard. However, if a faster, cheaper, more viable technique for evaluating asbestos exposure were available, it might reduce the amount of time and effort that is spent on counting asbestos fibers by the legally required method. For instance, surveying areas where asbestos aerosols are suspected to exist, would be made easier and faster by having a portable instrument capable of detecting fibrous dust. Such an instrument is currently being developed and constructed by the GCA Corporation under contract to NIOSH. For counting personal samples, it is desirable to have a method that is rapid, reproducible and equivalent to the legally required method. For OSHA compliance actions, it may still be necessary to use the present standard method, but for screening out samples that are obviously below the threshold values, and indicating those that are near or above these values, a less time consuming automated technique that is at least as sensitive as, and equivalent to, the visual counting method is needed. Sensitivity in context of fiber counting is here defined as the minimum diameter, 5 μm long fiber that can be observed.

The only currently viable alternative to the visual counting method by light microscopy is an electron microscopy method. This technique has much higher magnification and resolution available than light microscopy. In addition, it can be used in conjunction with electron diffraction and X-ray analysis to make the detection specific for asbestos. The drawbacks of electron microscopy for asbestos counting include the requirement of very expensive equipment, long analysis time using highly trained personnel, and considerable difficulties in sample preparation.

The standard method for counting asbestos fibers by light microscopy has a number of drawbacks as well. Typical counting times of 10 - 20 minutes per sample makes the technique tedious and time consuming. The precision of the results has been found to be poor (CV > 0.20)[2] when compared to other sampling and analytical procedures recommended by NIOSH (typically CV < 0.128).[10] The low precision has been related to lack of counter training and differences in counter motivation and eyesight. Apart from other physical, more quantifiable sources of variability, these human factors indicate that an automated instrumental method of asbestos counting is desirable. Two approaches have been taken in this regard. The first is to investigate the use of image analysis for counting light microscopy images of asbestos particles. This subject will be treated in another report. The second approach has been to investigate the use of light scattering as a means of detection of fibrous asbestos particles.

A considerable amount of experimental work has already been done on fiber detection systems. A contract is currently underway to design and construct a fibrous aerosol monitor that uses both light scattering and aerodynamic properties of fibers for their measurement.[9] Timbrell [11,12] has investigated light scattering from magnetically aligned fibers in liquid suspension and deposited on glass slides. This work has been done primarily as a means of rapidly identifying types of fibers based on their magnetic and light scattering properties.

What is proposed in this discussion is to develop the basis for the design and construction of a light scattering instrument that is capable of counting fibers on a filter in a manner analogous to and with a sensitivity equivalent to the present standard visual counting method. In brief, the proposed system involves the following: the filter is scanned by a focused laser beam; the light scattered from the surface of the filters is collected and analyzed electronically; light scattering signals indicative of fibers are counted, and; the results are displayed as a count of fibers per unit area of filter.

Theoretical Background

The scattering of light by acicular shapes has been studied theoretically by a number of investigators.[13] [14] A rigorous theory of scattering from particles has been developed by Mie, although for many important limiting cases, the Rayleigh-Gans approximation is adequate to describe the scattering function. Enough theoretical and experimental work has been done in this area to draw some general conclusions about light scattering from real fibers and the applicability of scattering to the detection and counting of fibers.

For the purposes of discussing the theoretical data from the literature, several parameters and assumptions relating to fibers have to be defined. In most cases, the length of the fibers is assumed to be infinite. Since the fiber size parameters are generally related to the wavelength of light being scattered, the approximation of infinite length is a reasonable limiting case. The wavelength of light, λ, will be assumed to be that of the He-Ne laser, i.e., $\lambda = 0.633$ µm. The size parameter α is defined as

$$\alpha = \frac{2 \pi a}{\lambda}$$

where a is the radius of the fiber.

The fiber is generally assumed to have a circular cross section and uniform density, although investigations of stratified and of elliptical fibers have been made.[14] The refractive index (m) of the fibers is an important parameter. The refractive index of asbestos fibers ranges from m = 1.55 for chrysotile to m = 1.71 for crocidolite. Since the fibers will be deposited on a slide or a filter, they will be in a plane perpendicular to the incident light. This fact greatly reduces the possibility of oblique incidence.

When a particle interacts with a light beam, some of the light is scattered, some of it is absorbed, and the remainder is transmitted. The extinction factor Qext indicates the sum of the factors for scattering Qscat and absorption Qabs. Some Qext curves for very long cylinders are given in Fig. 1.[15] The value along the abscissa is (m-1). If the particles are non-absorbing, then

$$Qext = Qscat$$

Oscillation in the extinction curves is due to interference between the diffracted and transmitted light. Any absorption by the fiber will subtract from Qscat and tend to smooth out the oscillations in the extinction curves. This is demonstrated in Figs. 2[16] and 3[17] where increasing amounts of absorption damp out the oscillations in the extinction curves. Since the extinction factors are a function of fiber diameter, this means that the scattering from long fibers will be approximately proportional to diameter at α larger than about 3. The overall result, as might be expected, is that the total scattering increases with increasing fiber diameter although not monotonically.

A primary point to be noted is that for infinitely long fibers, the scattering of light will occur only in a plane perpendicular to the fiber long axis and containing the incident light beam. This contrasts with spherical or roughly spherical particle scattering which occurs at all angles Θ about the incident beam. Referring to Fig. 4., where the incident beam is in the X direction, and the fiber is along the Z direction, scattering will only be observed in the X-Y plane. This fact has two important consequences for this discussion. First, this scattering pattern provides a distinctive means of differentiating between fibers and other less symmetric particles. Secondly, since all of the scattering occurs in one plane, the intensity in that plane will be higher than for nonfibrous particles and will decrease more slowly with distance from the scattering origin. For an infinitely long fiber, the scattering intensity decreases as $1/r$ where r is the distance from the fiber. For a spherical particle the intensity falls off as $1/r^2$.

Angular Dependence of Intensity

Up to this point, the discussion of light scattering has referred to the total scattering intensity. The scattering intensity as a function of the angle from the forward direction of the laser beam is also strongly dependent on particle size and refractive index. Fig. 5 [18] shows the dependence of back-scattering intensity at 180^0 as a function of fiber diameter. Fig. 6 [19] indicates the angle of the minima and maxima in plots like Fig. 5 as a function of fiber diameter. As can be seen from this type of information, it is unlikely that a scheme for precise sizing of even ideal fibers could be realized without using a great deal of analysis of the scattering data. Non-circular cross sections and non-uniformity of refractive index in real particles would make it virtually impossible to provide a rapid, accurate sizing technique by analysis of angular (Θ) distributions.

Polarization Effects

An additional effect that has been considered is the polarization of the light that is scattered from a fiber. A light beam can be considered to be polarized so that the electric field is either parallel or perpendicular to the fiber axis. All other angles of polarization can be treated as a linear combination of these two cases. The intensity for each case can be calculated and the polarization ratio is a sensitive function of fiber diameter and refractive index as shown in Fig. 7. This figure represents the attempt to theoretically fit some experimental data. It can be seen that small changes in either the refractive index or the fiber diameter cause relatively large changes in the polarization ratio as a function of scattering angle. This means that for well defined data, a theoretical fit can be obtained, but as seen for the angular intensity functions in the previous section, it would be very difficult to obtain rapid, accurate, size determinations from the polarization effects due to scattering from real fibers.

Figure 1 shows calculations of extinction coefficients for the electric field vector ($\overline{E}$) parallel to the fiber (case I) and perpendicular to the fiber (case II). It can be seen that for small ρ the ratio of parallel/perpendicular is large. Thus, small diameter fibers are detected more efficiently if the $\overline{E}$ polarization is parallel to the fiber axis. Under the appropriate circumstances, this factor could be used to reduce spurious light levels and increase sensitivity.

Spherical and Amorphous Particles:

Light scattering from spherical particles have been investigated more extensively than from other types of particles. The shape symmetry of any particle is reflected in its scattering pattern. For spherical particles, the high degree of symmetry results in scattering intensities that are constant for all Θ. The scattering intensity as a function of Θ is similar in many respects to the intensity function of fibers. As a matter of fact, a fiber can almost be considered a long chain of spherical particles. The scattering intensity is cancelled by destructive interference in all directions except in the X-Y plane. Particles, or clusters of particles, with a high degree of symmetry will have scattering patterns that have a similarly high degree of symmetry. This will be indicated to some extent by the scattering patterns of two or more spherical particles in the experimental discussions. The main point here is that the shape and symmetry of a particle is reflected in its scattering pattern. Hence, amorphous particles of ill-defined shape will have an ill-defined scattering pattern.

Discussion of Theoretically Expected Scattering

To understand the implications of the previous discussions of the theoretical predictions of scattering from fibers, it is convenient to set up the experimental situation to be implemented and to determine what observations might be expected. The experimental setup involves a long, straight fiber of arbitrary diameter on a glass slide. A narrow He-Ne laser beam impinges on the fiber perpendicular to the plane of the glass slide. The scattering is observed in the forward direction, i.e., at $\Theta<90^0$.

In actual measurements, the sum of the light intensity of some range of forward angles would actually be detected, e.g. $30^0 < \theta < 70^0$.

The total scattered intensity for the fiber is concentrated in a plane perpendicular to the fiber axis. The width of the scattering plane is essentially determined by the laser width. The intensity of scattering will generally decrease with increasing , but not monotonically.
To predict the detected intensity, the scattering function must be integrated over the range of angles of 30^0 to 70^0.

This integrated value will be proportional to the fiber diameter over limited ranges, by analogy to scattering from spherical particles. In general, the detected intensity will increase with increasing fiber diameter. For asbestos fibers, the imaginary part of the refractive index, or the absorbance of the fiber, is small. If the absorbance were large, this would decrease the scattering intensity and hence decrease the sensitivity of detection.

It may be noted that what one sees in a microscope when observing small fibers is the extinction of light by the fibers. The fiber is seen as a dark area where light has been removed by absorption and scattering. Since it is proposed to measure the scattering intensity for fibers with small absorbance, this process is quite analogous to the visual process of observing the fibers. It should therefore be possible to determine a threshold scattering intensity that agrees quite well with the sensitivity limit of the visual microscope technique.

Problems with Real Fibers

The above discussion has treated fibers primarily as long, straight single cylinders. When counting industrial hygiene fiber samples, one encounters many non-ideal fibers. Especially in the case of chrysotile asbestos, straight fibers are more the exception. The length of the fibers and the degree of overlapping and clumping varies considerably.

The straightness of the fibers presents a severe problem. If the laser beam intersects a curved portion of a fiber, the light will no longer be scattered into a plane, but into a range of angles about the plane, where this range of angles will be proportional to the curvature of the illuminated portion of the fiber. Thus the primary distinctive feature of the ideal fiber scattering pattern is degraded or destroyed. This problem can be overcome to some extent by focusing the laser beam to a small diameter, e.g. about 5 μm. Therefore, the curvature seen by the light beam is smaller and the fiber scattering pattern is not degraded as severely as demonstrated in Fig. 8.

One of the assumptions made for the purposes of calculating the light scattering behavior is that the fiber is infinitely long or at least very much longer than the wavelength of impinging light. This is in fact a fairly reasonable assumption since the fibers to be detected are longer than 5 μm and the wavelength of the light is 0.6 μm. A greater problem is encountered in the definition of a fiber, i.e., the minimum acceptable aspect ratio. As the aspect ratio decreases, the scattering pattern approaches that of a cubical or spherical particle. The point at which the scattering pattern is no longer sufficiently distinctive to allow its classification as a fiber must be determined experimentally. However, from observations of real fibers, it has been noted that this cutoff point is about 10:1 length to diameter or larger. This does not coincide with counting rules stated above which specify a greater than 3:1 aspect ratio.

In the cases of intersecting fibers or of more than one fiber in the laser beam at a time, each fiber will have its own scattering pattern. Thus crossed fibers or clumps are not a problem unless the fibers happen to be connected. This problem is probably not too important except in heavily concentrated samples, since the probability of having more than one fiber in a 5 μm beam area at any one time should be small.

This needs to be verified experimentally. The simplest treatment of the crossing or clumping problem would be to designate each observation of two or more simultaneous fiber scattering patterns from a single point as a single fiber. This will decrease the count rate somewhat, but probably not enough to be significant.

Experimental Information

A light microscope was modified as in Fig. 9 to use a 4 mW He-Ne laser as a light source. The laser beam passed through a spatial filter to obtain a collimated beam of more uniform cross section. The beam was then focused on the surface of the glass slide on which particles to be detected were placed. Passing the beam through a glass slide with an acetone cleared mixed cellulose ester filter reduces the intensity of the beam by approximately 5%. Most of this reduction in beam intensity is due to reflection from the surfaces rather than to scattering. Observation of particles in the light beam was accomplished by using the microscope objective and eyepiece assembly in its normal mode, and projecting the image onto a screen. A display of the scattering pattern was effected by removing the objective and eyepiece assembly and placing a ground glass screen approximately 15 mm above and parallel to the microscope slide. The main beam was intercepted by a blackened disc on the screen and the scattering pattern was observed through the screen.

The slit of the laser beam produced power densities at the focal area of about 30 μm diameter on the order of 5 Watts/mm. The high beam intensity was useful for visually observing the scattering patterns, since the scattering intensity is proportional to beam intensity at the surface of the slide. No damage to the filter surface was observed at these power densities, but this effect limits the laser power and the degree of slit that can be utilized.

The minimum length fiber to be observed is 5 μm. Asbestos fibers are composed of parallel aggregates of individual fibrils. These fibrils of asbestos are approximately 0.03 μm diameter. The cross sectional area for a fiber 5 μm by 0.03 μm is 0.15 μm^2. With a power density at the fiber of 5 $\mu Watts/m^2$, the fiber will scatter on the order of 0.5 μWatts of power. This power level can readily be detected by a photomultiplier. The light microscope can be used to detect fibers as thin as about 0.1 μm diameter. This means that sufficient light is scattered by all fibers that are to be detected if the fiber pattern can be differentiated from background and other interferences.

The resolution of the microscope when set up in the manner described above was relatively poor for several reasons. First, the wavelength of the He-Ne laser light is longer than the average wavelength of the white or the filtered light normally used in a light microscope. The resolving power is inversely proportional to the wavelength, so the use of 0.63 μm radiation reduces the resolving power by about a factor of two. The limiting numerical aperture (N.A.) of a microscope is determined both by the N.A. of the light condenser (spatial filter) as well as of the objective. In this case the objective N.A. was 0.65 but the effective condenser N.A. was only 0.02. The average of these two numbers can be used to estimate the resolving power. When the resolving power is defined as the radius of the diffraction disc around a particle, then two points may be resolved when they are separated by

$$d = \frac{0.61 \lambda}{N.A.}$$

The value for d, the laser illuminated microscope, is then about one micrometer. Particles smaller than about one micrometer could not be seen, so that this represented the available resolving power. However, it was possible to draw some conclusions from the larger particles and fibers that were observed.

The small N.A. essentially eliminates a problem common to the light microscopic method--namely, a small depth of field. At an N.A. = 0.02, the beam diameter remains within 120% of its minimum value over a distance of 5 times the minimum diameter. Even with a nominal beam diameter of 5 μm at the focal point, the beam diameter will be less than 5.5 μm over a depth of 25 μm. In comparison, the depth of field for an asbestos counting microscope is typically less than one μmeter.

Photographs (Figs. 10-20) were taken of some representative scattering patterns on the ground glass screen. The pictures were taken with a single lens reflex camera. The high contrast film produced some loss of the fainter portions of the patterns.

Spherical particles, glass fibers, asbestos fibers, and amorphous particles were observed with the experimental apparatus. Of these, the glass fibers and the amphibole asbestos fibers gave the clearest fiber-like scattering responses because they consisted largely of straight fibers. Chrysotile asbestos also gave fiber scattering patterns, but it was not possible in most cases to observe the particle that gave the response because of the limited resolving power of the microscope and the small diameter of the chrysotile fibers.

Figure 10 shows the scattering pattern observed for a single spherical 6 μm latex sphere.[21] There is a series of rings that represent the maxima in the scattering function as a function of Θ. Figure 11 shows the scattering pattern of two of these 6 μm spheres that are touching. Fig. 12 shows the pattern for four spheres in a straight chain. Note that the intensity is reduced in the direction along the axis and is starting to concentrate in the perpendicular direction. Some other non-linear arrangements of spheres produce interesting patterns. For instance, three spheres packed in a triangle produce the hexagonal pattern in Fig. 13. Four spheres in a T shape give the pattern in Fig. 14.

Glass fibers were looked at because they could be readily seen with the microscope (diameter of fibers typically 1 - 5 μm). The fibers were also relatively straight. Fibers greater than about 10:1 length to diameter had a sharp line pattern as indicated in Fig. 15. As the aspect ratio decreased, the scattering line broadened in ϕ and the intensity tended to concentrate in the near forward scattering direction.

Fig. 16 shows the pattern from a fiber with about a 10:1 aspect ratio. This gave a distinct pattern, but was relatively broad. A number of scattering patterns indicating a fiber were observed, but the particle causing these patterns could not be seen under the microscope. These were probably due to fibers too thin to be seen with the low N.A. microscope. Since scattering patterns from fibers presumably less than two μm in diameter could be seen visually, there is a good probability that instrumental detection, i.e. with a photomultiplier, will allow considerably higher sensitivity for detecting fibers than by the standard visual light microscopy method.

A major problem that has only been mentioned briefly up to this point is that of fiber curvature. The degree of curvature of a fiber in the region of laser illumination on the slide is translated directly into an increased angular spread of the scattering pattern. Hence, it is necessary to keep the beam diameter at the surface of the slide as small as possible, so that the curvature seen by the light beam is as small as possible. The optimum value of the beam diameter may be about 5 μm although this will have to be determined experimentally. This effect is indicated in Fig. 8, where the angular spread in the scattering pattern is smaller with a more tightly focussed laser beam.

There are other types of particles and structures that may produce a scattering pattern equivalent to that from one or more fibers. For instance, the scattering pattern from the straight edge of a large particle or an object larger than the beam size such as a coverslip edge will produce a scattering pattern as indicated in Fig. 17. This is essentially half of a fiber scattering pattern. A rectangular screen mesh will produce two fiber light scattering patterns at right angles if a sufficient portion of the mesh is illuminated. The only other structure that might produce a fiber like scattering pattern is a cubic crystal such as NaCl or larger crystals with straight edges. Particles of NaCl have been investigated and, in general, only very large particles (>30 μm) gave reasonably sharp scattering patterns. As the particles became smaller, the scattering patterns tended to become more diffuse. Some examples of this are shown in Figs. 18 and 19. This indicates that it may be necessary to remove the very large particles that may exist in the industrial atmosphere by a preselection device such as a cyclone. This would remove, at the sample collection stage, most of the possible interferences to fiber scattering detection.

A situation has been noted where fiber like scattering patterns were produced with acetone cleared filters under coverslips. Evidently, under certain conditions, fracturing between the surface of the filter and the coverslip occurs resulting in long thin fracture marks. These marks produce fiber scattering patterns.

The overlapping of scattering patterns from amorphous particles and from fibers does not cause a significant problem unless, of course, the fiber is completely or almost completely covered by the amorphous particle. For instance, in the situation where a fiber sticks out of an amorphous particle, the fiber can readily by detected if the portion sticking out meets the fiber criteria of length and aspect ratio.

When more than one fiber is illuminated by the laser beam, each fiber will produce its own scattering pattern as indicated in Fig. 20. Hence, fiber crossing does not present a problem in detection in a light scattering system. However, the situation is complicated by the occurence of fibers with frayed ends which are normally counted as a single fiber. This problem of how to treat the observation of more than one fiber simultaneously will be discussed under the section on counting strategy below.

In order to standardize the response of a light scattering instrument, it is necessary to have a calibration particle or image that can be widely used. The lines on a stage micrometer may be used for this purpose. The scattering from a single line on standard 0.01 mm stage micrometer is shown in Fig. 21. The pattern is well defined and should be quite uniform from micrometer to micrometer from the same manufacturer.

From what has been predicted theoretically and from what has been observed, it seems unlikely that any industrial aerosol particles will produce fiber scattering patterns in significant numbers except fibers. The major problem in detection is the occurrence of fibers with a high degree of curvature. This may be alleviated to a large extent by the use of as small a beam size as possible. However, there may have to be a statistical factor figured into the results of a fiber count to take this aspect into consideration.

Proposed Instrument

The previous discussion indicates that it may be possible to construct an instrument capable of counting fibers on a filter. The filter with sample is prepared in the same manner as recommended in the USPHS/NIOSH method or in an equivalent fashion. The problem then is to construct an instrument to scan the area of the filter in such a manner as to get an exact or an accurately estimated count of the number of fibers on the filter. There are probably any number of instrumental ways of accomplishing this goal, but the following includes a number of ideas developed in the course of this investigation that may help in the final design of such an instrument. There are several basic components: the light source, the optics and detection system, the scanning mechanism, and the data analysis system.

The Light Source

In terms of the requirements of light intensity, small diameter focusing, and stability, a laser is the obvious choice as a light source. Based on the ease with which larger fibers could be detected visually with a 4 mW He-Ne laser, a one milliwatt laser would probably be adequate in the final instrument. From the viewpoint of cost, availability, safety, and power consumption, the 1 mW He-Ne laser is the obvious choice. It is doubtful that significant advantages would be accrued by using a shorter wavelength laser which might give greater sensitivity.

The Optics

The laser beam must be transmitted from the source and focused into a well defined beam of uniform intensity at the surface of the filter. The scattered light must then be gathered and detected.

The light focusing elements can consist of a conventional spatial filter with one lens focusing onto a pinhole and the second lens focusing at the surface of the filter. Keeping the N.A. at a minimum will increase the depth of focus at the surface, so that exact determination of the focal plane is not a critical factor. The pinhole allows the selection of the most intense and uniform part of the beam. The size and positioning of the pinhole is critical in obtaining maximum intensity and the desired beam size of approximately 5 μm diameter at the filter surface.

The scattered light to be detected is scattered between angles of Θ' of approximately 30° and 70°. At smaller angles, the scattering from other particulates and from the filter itself gives too great a background and at greater angles, light collection is physically limited by the planar surface of the filter and perhaps by the coverslip. With a coverslip on the filter, internal reflection at the surface of the glass limits the angle at which the scattered light can escape in the forward direction. The coverslip may be necessary to ensure the planarity of the filter surface.

One possibility at this point is to have the detector look at the scattered light through a slit and move the detector around the laser beam axis to detect fiber scattering patterns. This arrangement was tried with a photomultiplier/electrometer detection system. The fiber patterns were recorded as two approximately equal signals 180° apart in Θ. The angular scan was produced by rotating the microscope stage. This detection/scan arrangement is too awkward to be useful, especially at the speed necessary to cover a reasonable portion of the filter. A more useful geometry would be to collect the scattered light with an elliptical mirror as shown in Fig. 22, where the fiber is at one focal point of the ellipse and the detector is near or at the second focal point. A light trap prevents the forward scattering and main beam from reaching the detector. A rotating chopper blade with slits (Fig. 23) is interposed along the optical path before the detector to make the detection specific for fibers. The reasons for using three slits rather than one are to balance the rotating mass of the blade, and to increase the detection frequency and hence the rate at which the filter can be scanned. Any higher odd number of slits could also be used if necessary. With three slits, the fibers are detected as six pulses 60° apart during a single rotation of the blade. The angular opening of the slits are optimized experimentally to accommodate an acceptable minimum fiber length to width ratio and to accommodate slightly curved fibers. In this configuration, a single stationary detector may be used. By using a high speed rotating blade at about 100,000 rpm,[22] a total of 10 pulses can be obtained in one millisecond (ms), allowing for adequate integration of the pulses and good signal to noise. These numbers will depend on the sensitivity and time response of the photodetector.

An alternative to the detector-slit geometry shown in Fig. 22 is the one shown in Fig. 24, where the slits are replaced by mirrors and the rest of the chopper blade is light absorbing. This arrangement may allow a more isolated and convenient placement of the chopper motor.

If the beam spot size is 5 μm and the detection time is 1 ms, the fastest linear scan rate possible is 5 μm/ms. The rotating slit/detector arrangement may not be optimum and needs further development. Rather than using the slit arrangement described, it may be easier to use a fiber optic arrangement to achieve the same effect. A higher rate of detection of the scattering patterns would allow a faster scan of the filter.

Filter Scanning Mechanism

As discussed above, the scanning rate of the filter is estimated to be 5 μm/ms. This is obviously not fast enough to scan an entire area of a 37-mm filter in a reasonable length of time. With a fiber concentration of 2 fibers/cm^3, a 3 hour sample collected at 2 l/min will deposit approximately 5.40 x 10^5 fibers on an area of 960 mm^2. This results in 560 fibers/mm^2. If the filter is scanned for three minutes at the above rate with a 5 μm beam diameter, an area of about 4.5 mm^2 will be scanned, which will intersect 2500 fibers. This is an adequate number for high statistical precision and allows ample of leeway for measuring less concentrated aerosols or for taking smaller air samples.

The easiest scanning mode would be to have the laser beam stationary and move the filter. X-Y positioning platforms are readily available that are compatible with the positioning accuracy and speed desired. Two basic scan patterns can be used--a linear raster of scans or a spiral pattern starting from the center of the filter. The former scan pattern may be more versatile if the entire filter is not used or cleared, while the second scan pattern may be accomplished in a somewhat simpler fashion and gave a better statistical and spatial sampling of the filter surface.

Signal Analysis

The electronic signal analysis system is shown in Fig. 25. The signal output of the photodetector is fed into a pulse amplifier that eliminates slowly varying signals and only detects pulses from sharply defined fiber scattering patterns. Longer pulses are rejected. The pulses are then shaped and fed into an amplifier tuned to six times the blade rotation frequency. The output of this amplifier is detected and integrated to give a signal pulse each time the laser beam intersects a fiber. The minimum pulse width will occur when the fiber is perpendicular to the direction of scan. The pulse will be the longest when the laser beam intersects a fiber that is parallel to and lies along the direction of the scan. The next step is a threshold gate that allows only pulses of a minimum height to pass. This threshold is set to make the detection process equivalent in sensitivity to the visual microscopy method. The pulses are then counted and divided by the total area scanned to give the final result.

The signal analysis system described does not respond to more than one fiber detected at a time, i.e., it counts fibers crossing within the area of the laser beam as a single fiber. This also means that if two or more very fine fibers are detected simultaneously that would not have met the threshold criterion separately, they may be counted as a fiber. If the fiber size distribution is skewed to the extremely fine fibers, multiple fine fiber detection may contribute significantly to the fiber count. However, the fiber density on the filter is normally kept low enough by adjusting the air sampling time that this situation should not become a serious problem.

The final value that the instrument operator obtains can be expressed in either units of fibers/mm^2 or fiber/cm^3 if the sampled air volume is fed into the instrument. The most straightforward approach to obtaining this final result would be to incorporate a microprocessor into the instrument to serve as the "brains." The microprocessor can not only perform the final calculation to obtain the final result, but also control the movement of the stage, and coordinate the scan with the detection circuitry. In addition, the versatility of the microprocessor allows the use of more complex scan patterns and implementation of appropriate calibrations when necessary.

There are a number of factors that have to be taken into account when setting up the instrument as described. First of all, the intensity of the laser must be normalized or calibrated to give a scattering response signal that can be readily compared with other identical or equivalent instruments. This would take into account variations in output power between lasers, the beam area at the surface of the filter, losses in the optical system due to scattering and absorption, and the variation in detection geometry and efficiency. Once this calibration has been obtained, a threshold must be determined that makes the detection system equivalent in sensitivity to the visual microscopy standard counting method. Determination of this threshold must be made by comparison of experimental counting by the visual and instrumental methods. A third design variable that is related to fiber dimensions as mentioned previously is the angular width of the rotating slits. This slit width can be determined by observing the pattern for known aspect ratio fibers and setting the slit dimension to cut off at an appropriate aspect ratio. The aspect ratio finally used will likely be greater than 3:1 used in visual counting because such a small ratio would not allow the instrument to be sufficiently specific for fibers. A ratio between 5:1 and 10:1 may be more useful and provide better fiber discrimination. However, in some cases, such as tremolite and anthrophyllite, the asbestos samples may contain a significant portion of fibers in the 3:1 to 10:1 aspect ratio range.

In determining some of these design variables, it may be useful to approach the optimum values by using non-fibrous samples. For instance, a sample of amorphous particles could be scanned and the slit width widened until the number of "fiber" counts rose above an acceptable level.

Counting Strategy

Since the entire filter cannot be scanned in a reasonable time due to instrumental limitations, a strategy for choosing an appropriate procedure for scanning the filter surface has to be devised. The major factor that limits how close together points on the filter surface can be observed is determined by the maximum expected length of the fibers (for instance 100 μm). If points are taken continuously in a line scan, then a single fiber or the frayed ends of a single fiber can be detected more than once and give a spuriously high count. For the same reason, two scan lines cannot be closer than the maximum expected fiber length. If a line scan is used, then each time a fiber is detected, the signal from a certain distance (e.g. 100 µm) of the scan is blanked out to prevent counting the same fiber twice.

An alternative to line scanning is point sampling. In this type of scan, a grid of discrete points is scanned, with the points being at least 100 µm apart. The main disadvantage is the time it takes to mechanically move the slide from scan point to scan point. A significant increase in total analysis time will result from such an approach.

If the entire 37-mm filter is placed on the stage and cleared properly, the most representative sampling of the surface can be obtained using a spiral scan starting from the center. This pattern eliminates radial and angular bias that may be due to the configuration of the filter holder. Alternatively, a linear raster that covers the surface of the filter or filter section can be used. The spacing of the raster lines can be such as to cover the entire surface. For instance, assuming a scan rate of 5 µm/ms for a scan time of 3 minutes, the length of the scan is approximately 900 mm. The scan lines of the raster are then approximately 1 mm apart if the entire surface of a 37-mm filter is covered.

Each time a signal is observed, it is registered as one fiber. The probability of a pair of crossed or of several fibers occurring in the beam at one time is assumed to be negligible. When a fiber is counted, all following signals for a predetermined scan distance, such as 100 μm, are ignored. To obtain the final result, in fibers/mm^2, the number of signals observed (F) is divided by the product of the length of the scan (L) when signal was being accepted and the beam width, (B)

$$\frac{\#\ \text{Fibers}}{\text{mm}^2} = \frac{F}{L \times B}$$

If the scan is a fixed length (L_0) then

$$L = L_0 - F \times 0.1 \text{ mm}$$

to allow for the dead time in the scan because of fibers observed. This calculation is simple enough to be accomplished by the calculator chip or microprocessor used in the instrument.

Because of the many variables involved in the light scattering detection process, the final calibration of the instrument must be performed against qualified counters using the standard method. The precision and biases in both techniques will have to be balanced to come up with conversion factors for each type of asbestos so that the light scattering instrument can be effectively used as a screening and analysis device for industrial hygiene samples.

Conclusions

It is felt that an instrument based on light scattering from fibers can provide accurate and precise results for analysis of industrial hygiene samples of asbestos. The proposed instrument uses a physical basis for detection that is quite analogous to that in the present standard method. It can be automated to provide a rapid result with high statistical precision. There are unknown biases due to interference by other fibrous particles, to interference by nonfibrous particles, and to high degrees of curvature of some asbestos fibers, but appropriate calibration of the instrument should alleviate these problems.

The instrument as proposed contains many assumptions that remain to be investigated. The validity of these assumptions can best be probed by constructing a prototype based on the general outline presented and performing numerous measurements on the light scattering properties of individual fibers as well as of field filter samples as a whole.

Table I contains price estimates for the various components based on commercially available equipment. By in-house manufacture, many of these components can be reduced in price considerably. The price of the various items will depend heavily on the final approach taken in their construction. The final commercial cost of a fiber counting instrument should be under \$20,000 and possibly even under \$10,000. Considering the time savings in sampling counting, such a fiber counting instrument would present a cost effective approach to the analysis of industrial hygiene asbestos samples.

An additional technique that might be used is one proposed by Bartosiewicz[23] where half the sample is counted in an index of refraction medium that is the same as that of the type of asbestos being analyzed and half is counted in a different index of refraction medium. In the former medium, the asbestos will not scatter light and in the second it will, so that a qualitative as well as quantitative procedure can be set up.

In the same paper by Bartosiewicz , a suggestion for a closed face filter sampler is made that purportedly deposits aerosols uniformly over the face of the filter. Since a cyclone or an equivalent preselection device may be useful in removing interferences to fiber counting, such a sampler may be necessary to provide uniformly deposited filter samples. The evaluation of such a sampling train and other fiber sampling systems would be made easier with the automated fiber counter that has been proposed.

REFERENCES

1. NIOSH Criteria Document for Asbestos, (1972).
2. P. Baron, manuscript in preparation, to be published in Annals of Occupational Hygiene.
3. Occupational Safety and Health Standards 29 CFR 1910.93a, Federal Register, June 27, 1974 (also 29 CFR 1910.1001).
4. J. Lynch, H. Ayer, and D. Johnson, AIHA Journal 31 (1970), p. 598.
5. O. Menis, Proceedings of a Round Table on Thermal Analysis Techniques in Occupational Safety and Health. NIOSH Publication, April 16-17 (1975), p. 53.
6. A. Richards, Analytical Chem. 44 (1972), p. 1872.
7. Private Communication, Dr. Peter Eller, NIOSH.
8. Private Communication, Dr. Janet Haartz, NIOSH.
9. NIOSH Contract No. 210-76-0110. Development and Fabrication of a Prototype Fibrous Aerosol Monitor.
10. NIOSH Standards Completion Program, Contract 74-45.
11. V. Timbrell, Presented at IOEH Conference, Montreal, Oct. 29-30 (1973).
12. V. Timbrell, Microscope 20 1972, p. 365.
13. H. Van de Hulst, Light Scattering by Small Particles, John Wiley and Sons (1957).
14. M. Kerker, The Scattering of Light, Academic Press (1969).
15. M. Kerker, Ibid, p. 290.
16. M. Kerker, Ibid, p. 295.
17. M. Kerker, Ibid, p. 296.
18. M. Kerker, Ibid, p. 293.
19. M. Kerker, Ibid, p. 294
20. M. Kerker, Ibid, p. 308.
21. The latex shperes were manufactured by Dow Chemical Co. out of styrene divinyl benzene to have a diameter of 5.7 μm with a standard deviation of 1.5.

REFERENCES

22. P. Selzer and N. Yen, Rev. Sci. Instr. 47, 747, (1976), also:
R. Nemanich, S. Solin, and J. Doehler, Rev. Sci. Inst. 47, 741, (1976).

23. L. Bartosiewicz, AIHA Journal, June (1973), p. 252.

APPENDIX I

The number of fibers/μg for asbestos has been calculated using data from two sources. The first source is data from the Proficiency Analytical Testing (PAT) Program, [2] which used laboratory prepared samples sent to a number of different laboratories for counting. The PAT average is 1.3×10^5 fibers/μg. A study by Lynch, et. al.[4] indicated a range of $0.67 - 2.4 \times 10^4$ fibers/μg. Thus, for a sample collected at the recommended rate of 1.5 lpm, for a reasonable sampling period of 6 hours, the mass of sample at the standard level of 2 fibers/cm^3 can range from 8 to 160 μg.

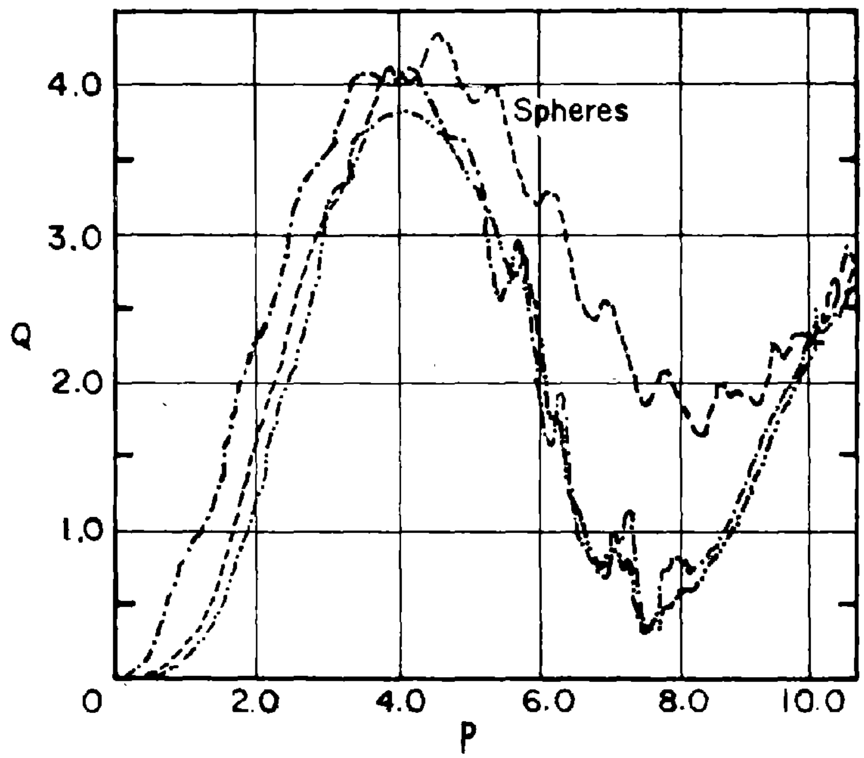


Fig. 1. Scattering efficiency of infinite cylinder (Q_1 and Q_2) with m = 1.48 compared with scattering efficiency of sphere (Q_{sca}) with m = 1.486. The abscissa is $\rho = 2\alpha(m - 1)$ (Farone et al., 1963) (Used with permission of author and publisher).

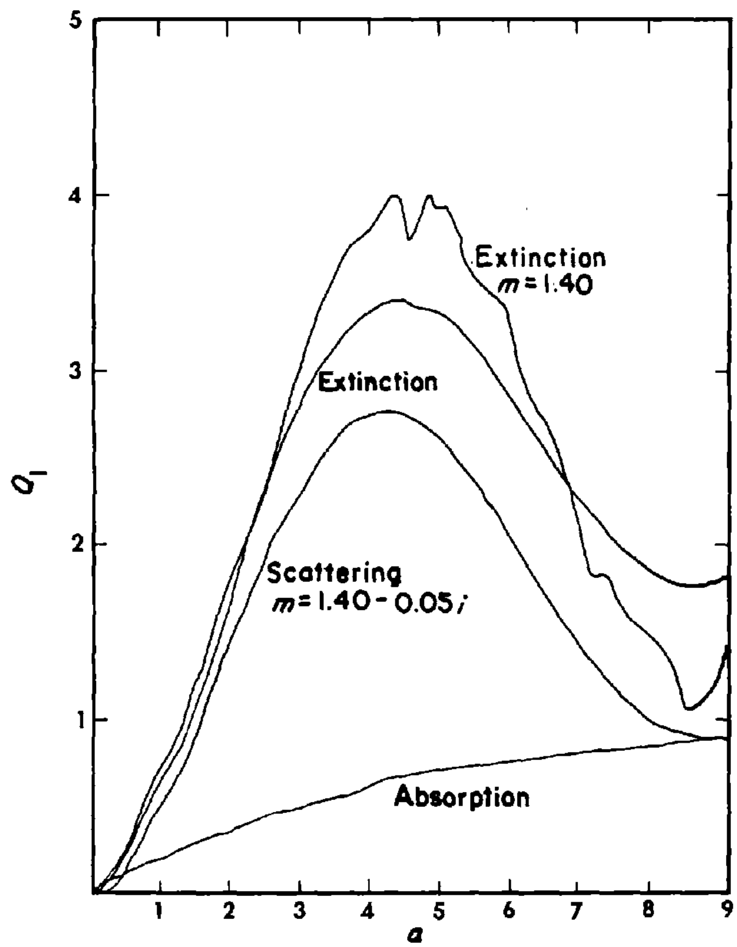


Fig. 2. Extinction efficiency Q_1 for m = 1.40 and the extinction, scattering and absorption efficiencies for m = 1.40 - 0.05i. (Kerker, 1969). (Used with permission of author and publisher).

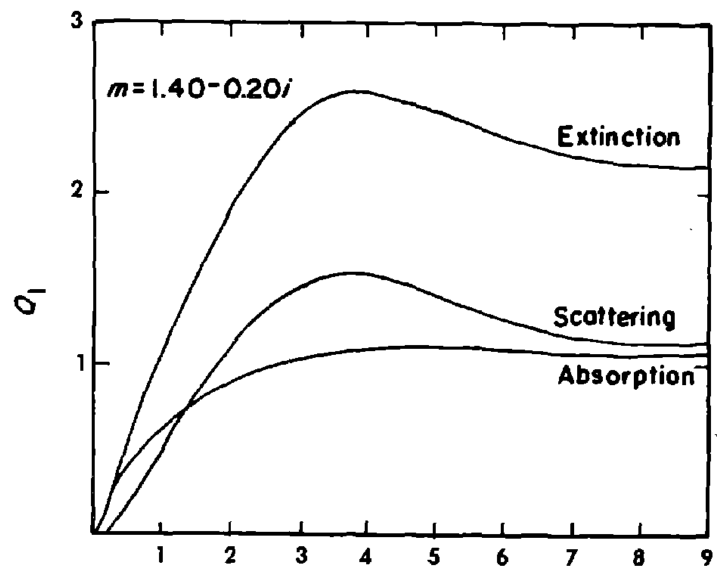


Fig. 3. Same as Fig. 2 for m = 1.40 - 0.20i. (Kerker, 1969). (Used with permission of author and publisher).

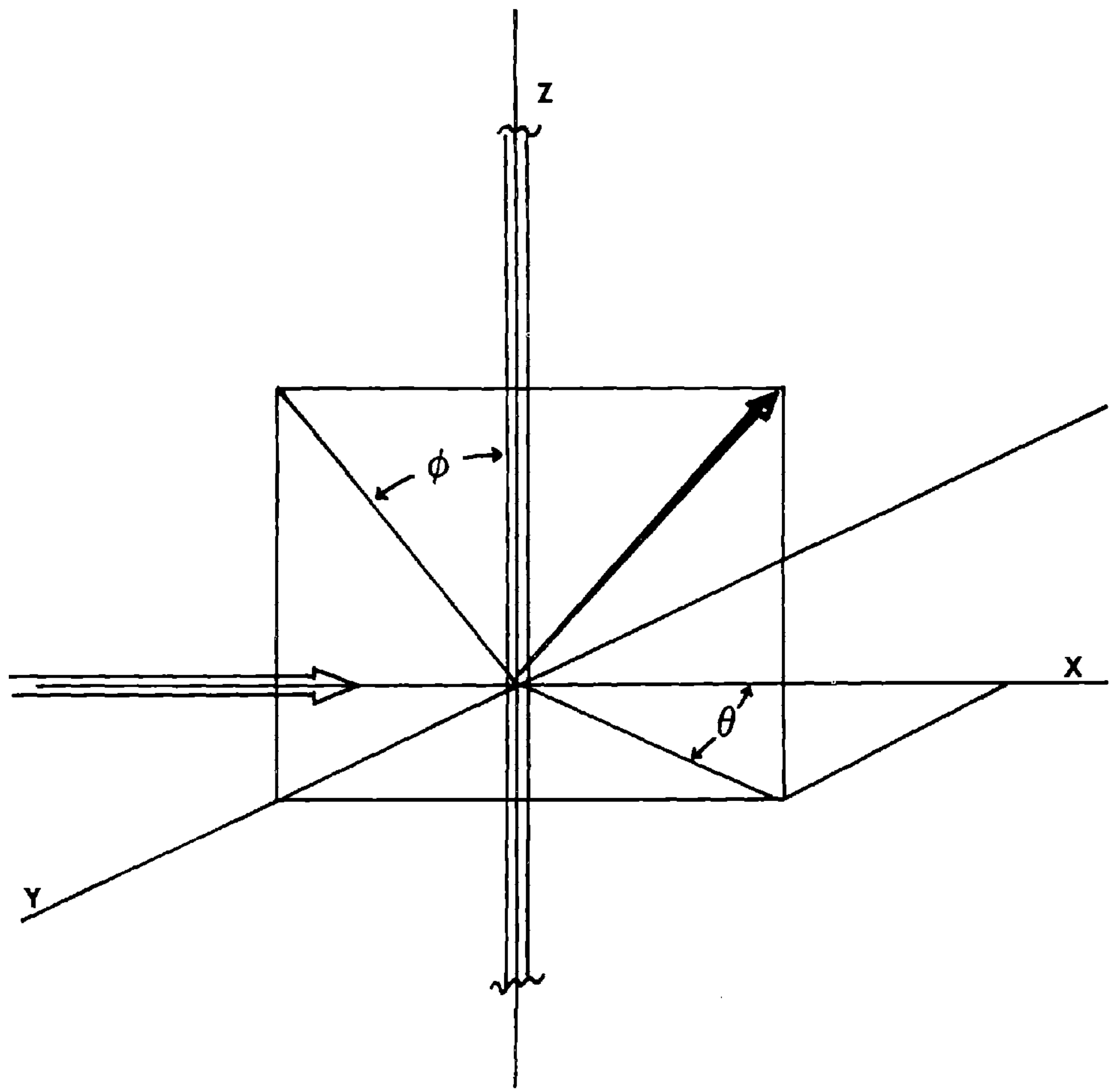


Fig. 4. The fiber is oriented along the Z direction and the light beam is in the +X direction.

The angle away from the +X direction Θ and the angle away from the +Z direction is ϕ.

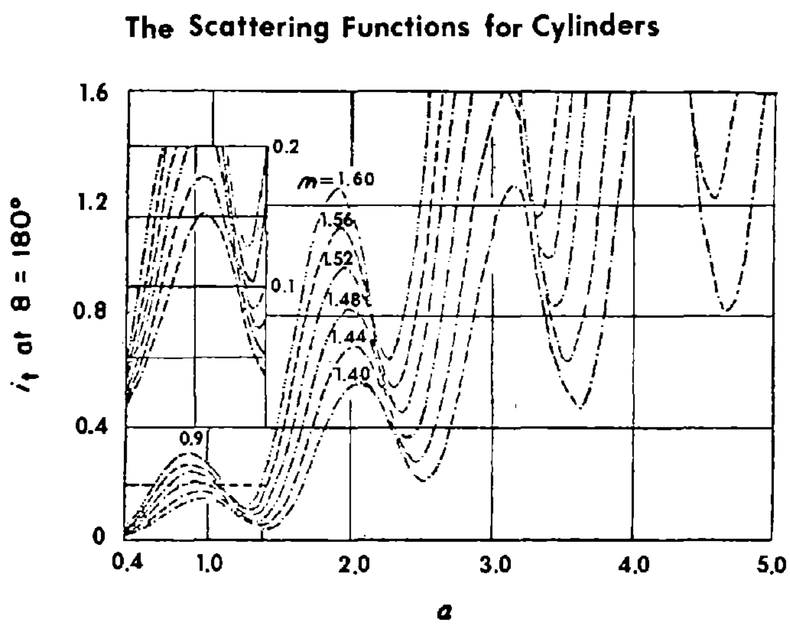


Fig. 5. Variation of angular scattering function i_1 with α at $\theta = 180^0$ for m = 1.40, 1.44, 1.48, 1.52, 1.56, and 1.60. The values of the maxima near α = 3.0 are 1.25, 1.60, 1.97, 2.32, 2.76 and 3.08, respectively, and those near α = 4.0 are 2.06, 2.70, 3.36, 4.04, 4.68 and 5.28, respectively. (Farone et al., 1963). (Used with permission of author and and publisher).

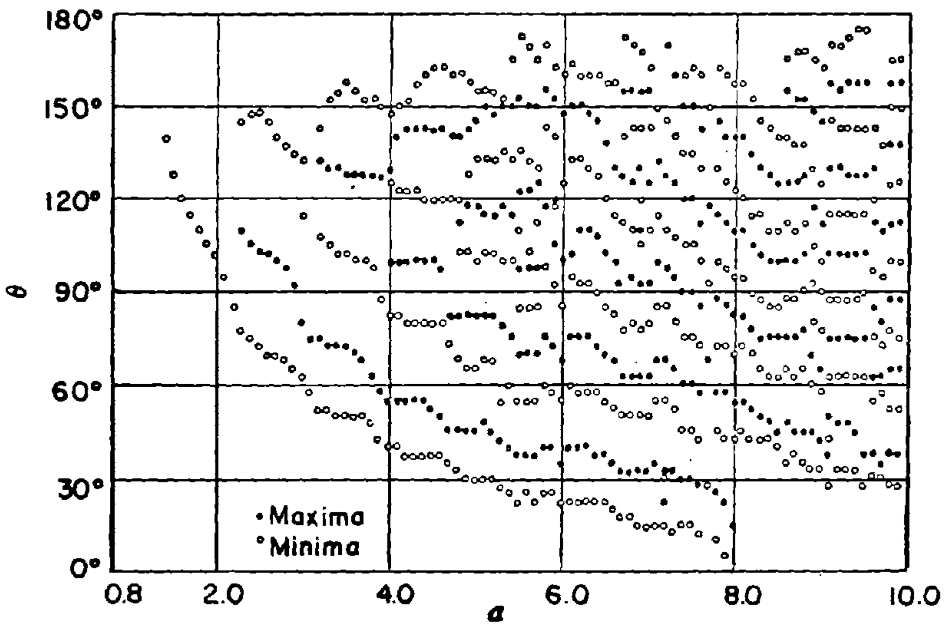


Fig. 6. Angular locations of the maxima (•) and minima (o) of i_1 for m = 1.46 (Farone et al., 1963). Used with permission of author and publisher).

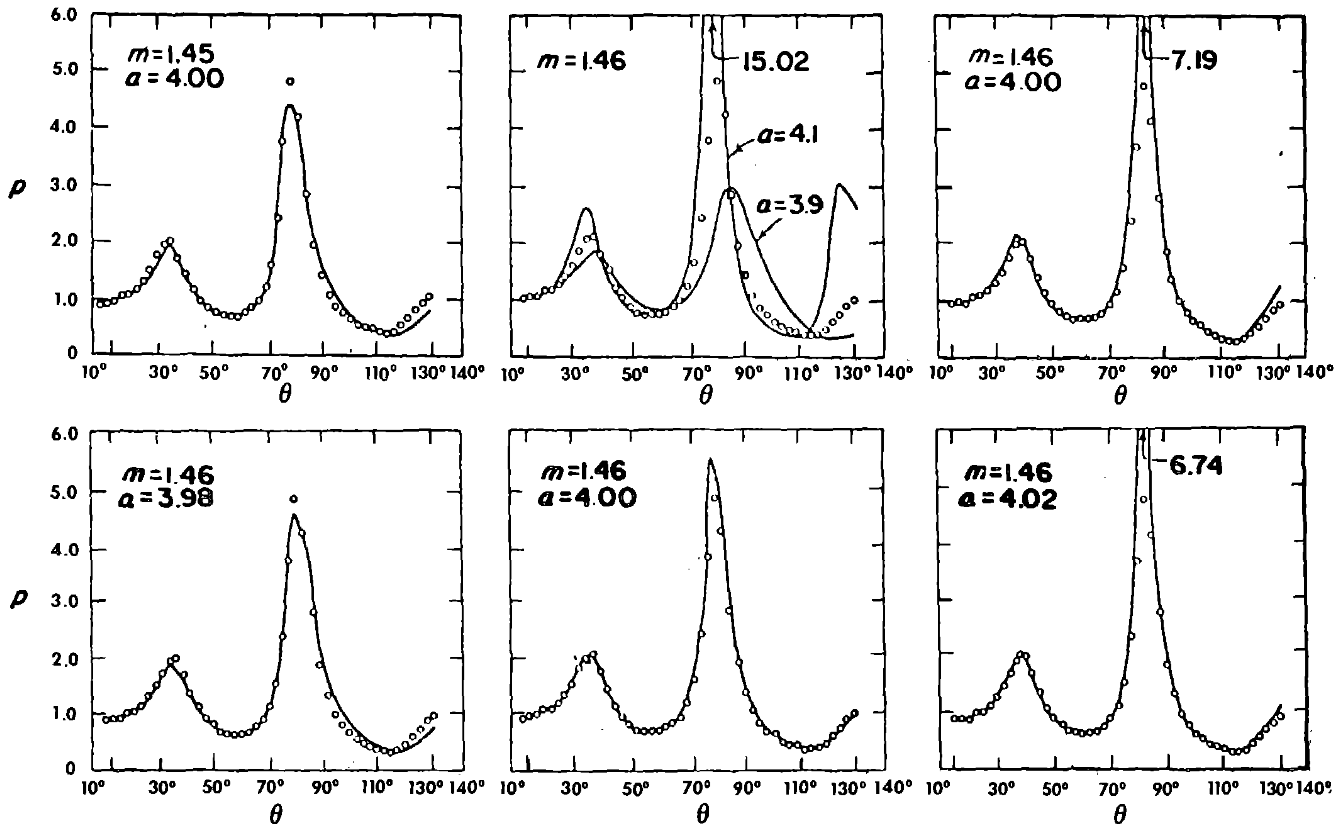


Fig. 7. Polarization ratio ($\rho = I_2/I_1$) plotted against θ for a Pyrex cylinder illuminated at perpendicular incidence. Comparison of experimental results (circles) and theory (smooth curves) for a silica fiber at $\lambda = 0.546\ \mu$ (Farone and Kerker, 1966). (Used with permission of author and publisher).

FIGURE 8

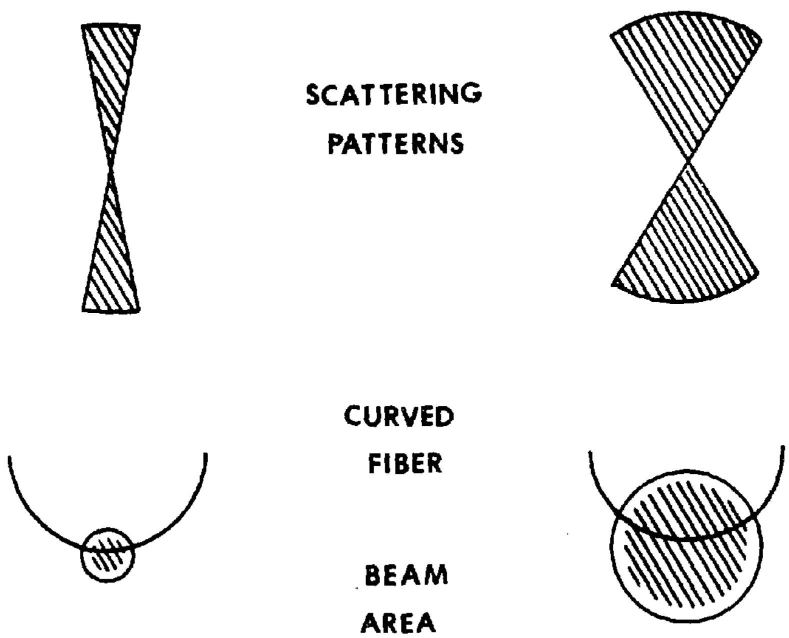
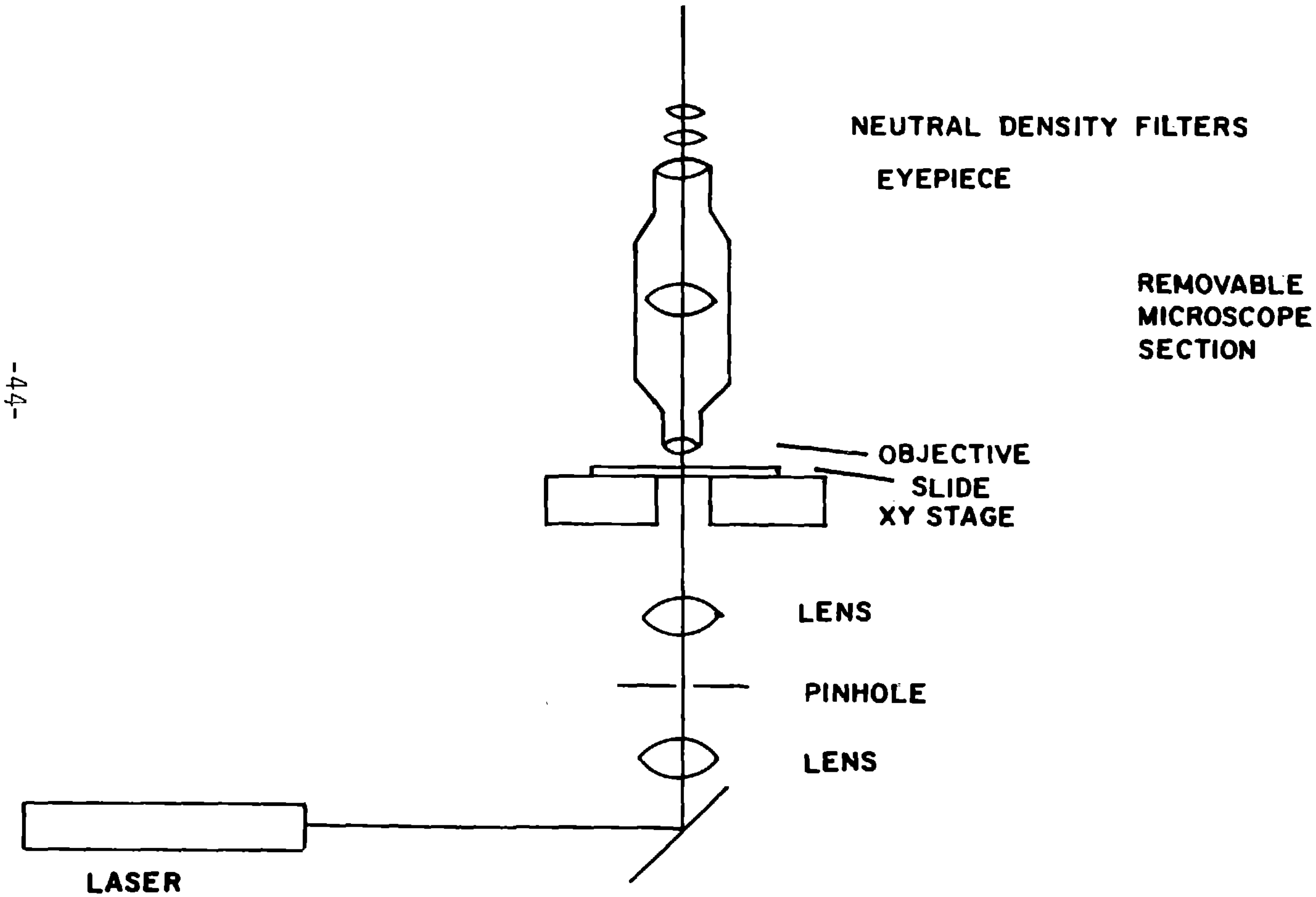


FIGURE 9


NEUTRAL DENSITY FILTERS
EYEPIECE
REMOVABLE MICROSCOPE SECTION
OBJECTIVE
SLIDE
XY STAGE
LENS
PINHOLE
LENS
LASER

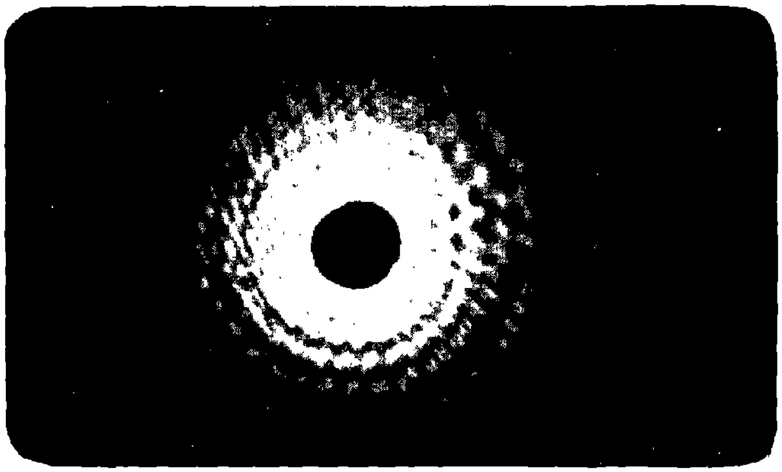


FIG. 10 Scattering Pattern from a Spherical Particle

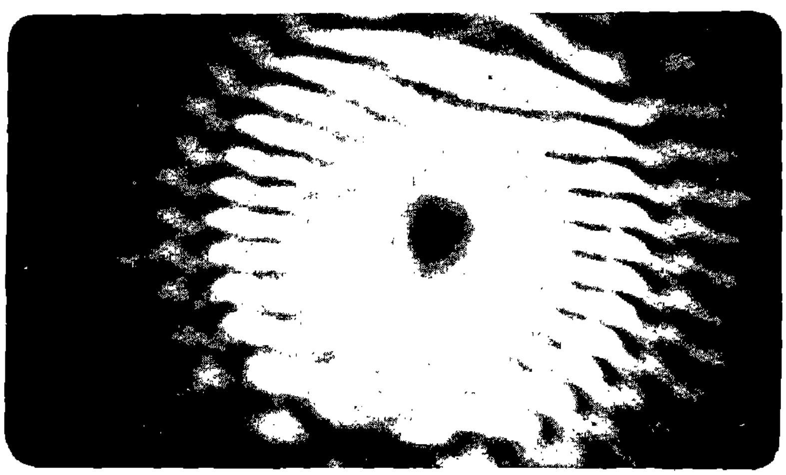


Fig. 11 Scattering Pattern from Two Spherical Particles

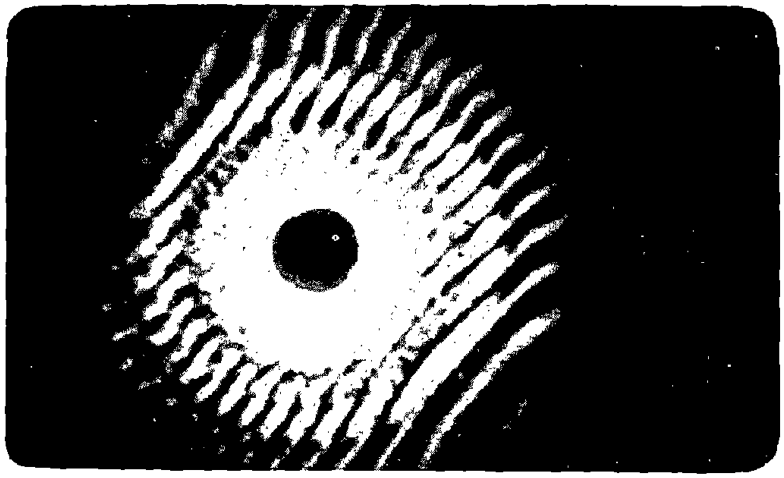


Fig. 12 Scattering Pattern from Four Spherical Particles in Line

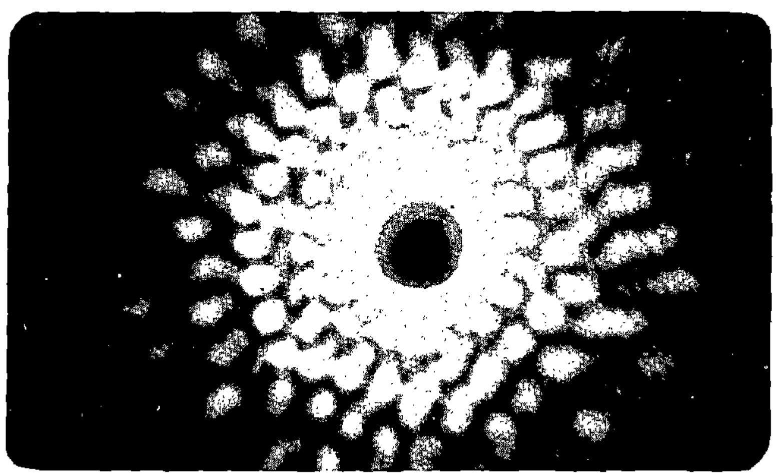


Fig. 13 Scattering Pattern from Three Spherical Particles in a Triangle

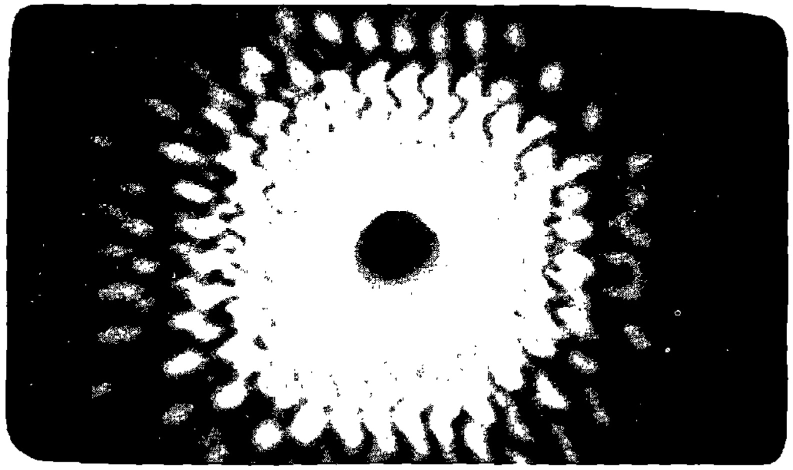


FIG. 14 Scattering Pattern from Four Spherical Particles in a T Arrangement

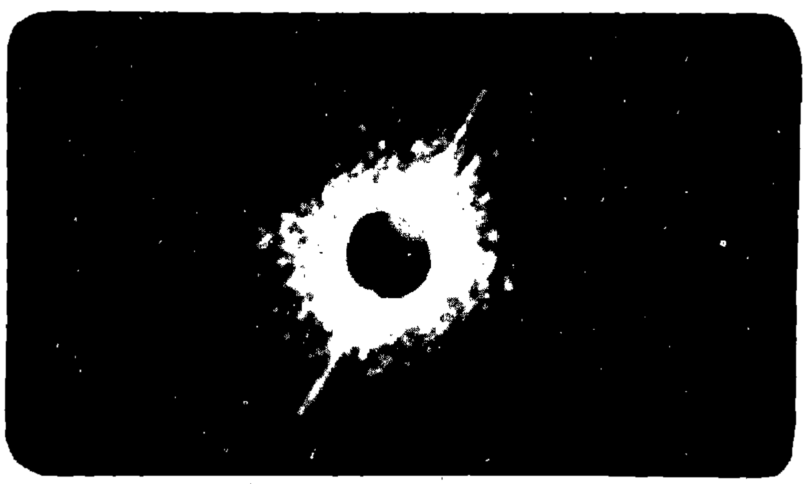


FIG. 15 Scattering Pattern from a Long Thin Fiber

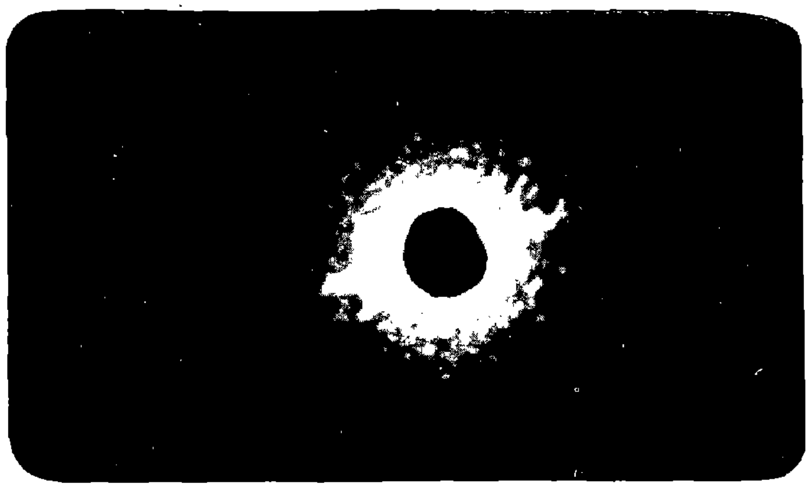


FIG. 16 Scattering Pattern from a Fiber with about 10:1 Aspect Ratio.

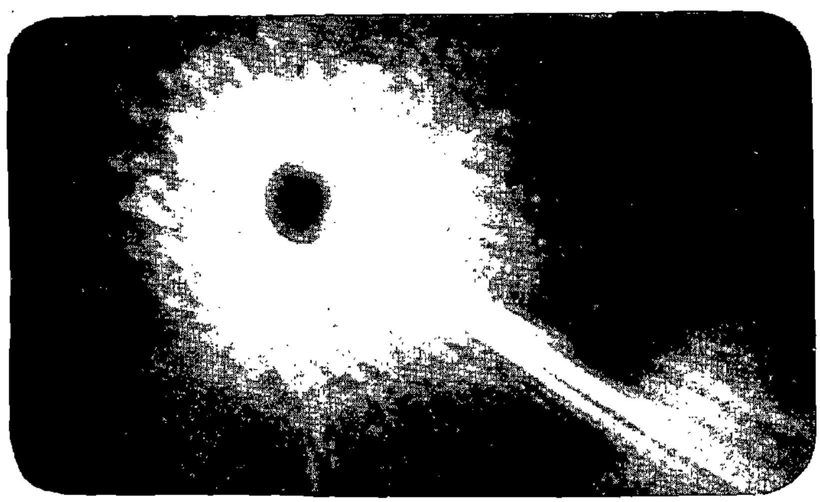


FIG. 17 Scattering Pattern from the Coverslip Edge.

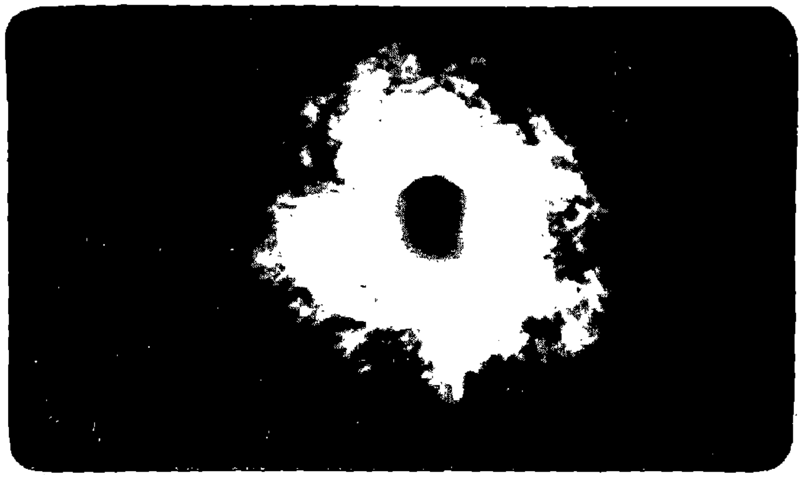


FIG. 18 Scattering Pattern from Amorphous Particle.

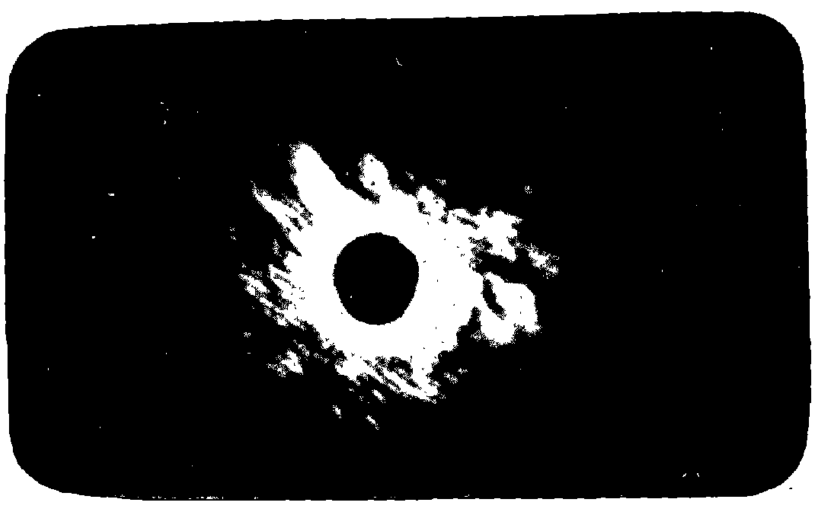


FIG. 19 Scattering Pattern from Amorphous Particle.

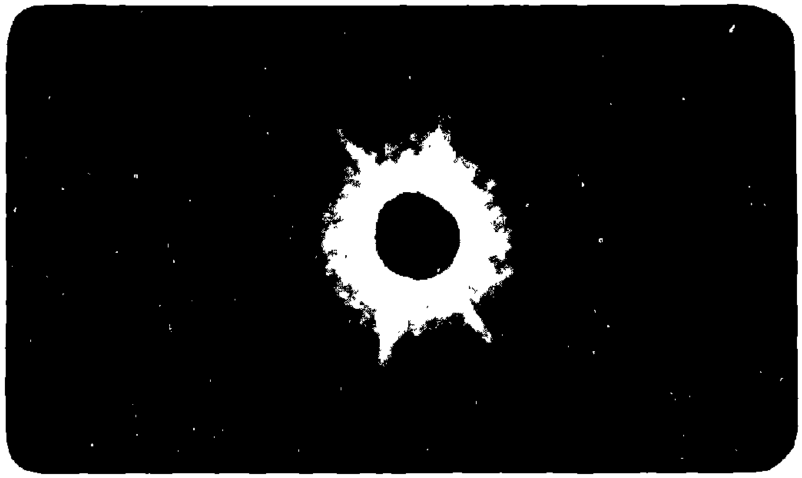


FIG. 20 Scattering Pattern from Two
Overlapping Fibers.

FIG. 21 Scattering Pattern From Stage
Micrometer Line.

FIGURE 22

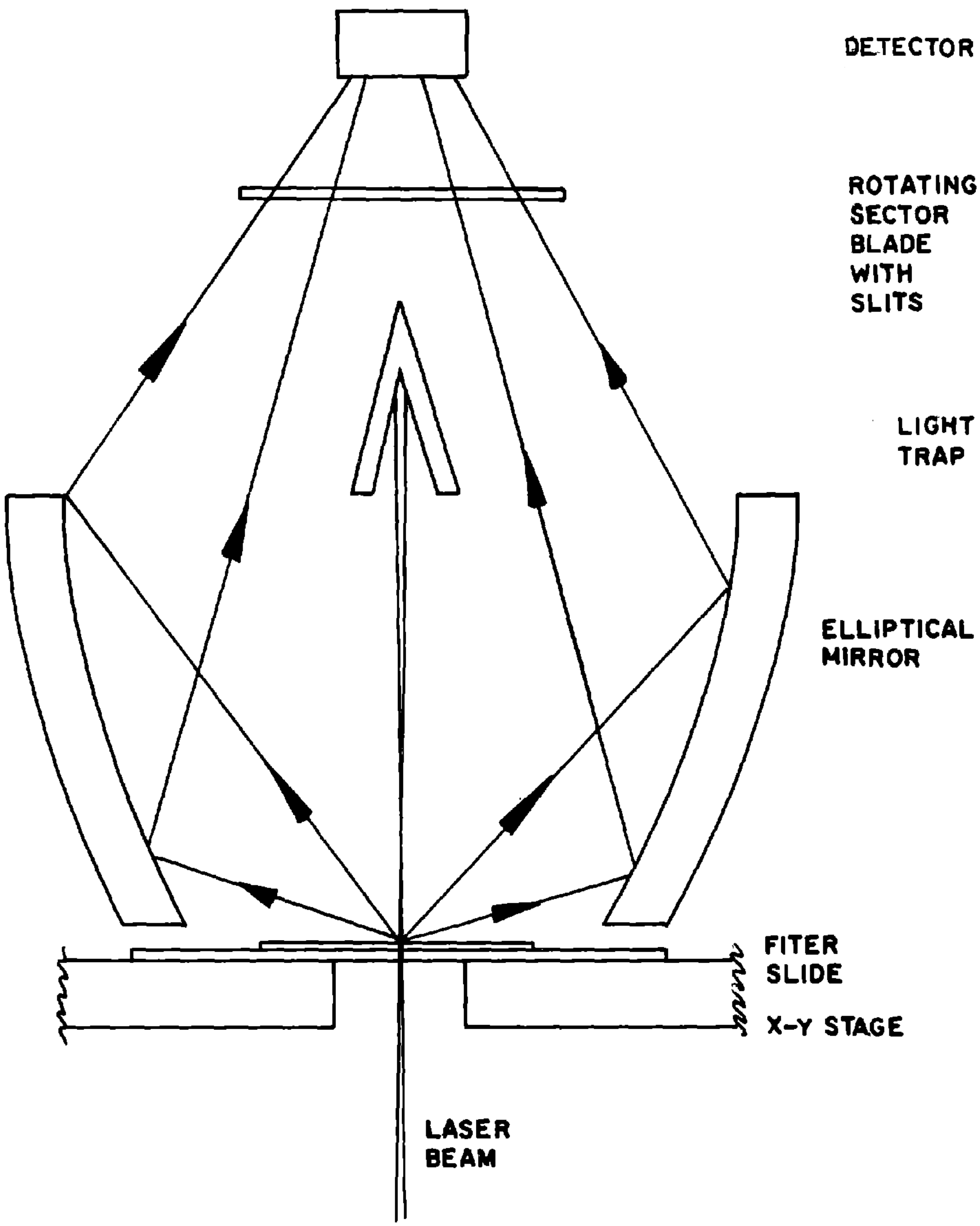

DETECTOR
ROTATING
SECTOR
BLADE
WITH
SLITS
LIGHT
TRAP
ELLIPTICAL
MIRROR
FITER
SLIDE
X-Y STAGE
LASER
BEAM

TABLE 1.

Component Price Estimate

Laser	200
Spatial Filter	700
Motorized X-Y Stage	1000
Electronics	1000
Optics	300
Detector and Chopper	800
Miscellaneous	1000
Total	5000

☆ U.S. GOVERNMENT PRINTING OFFICE 1978— 757-141/6768