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Small-Angle Light Scattering for Analysis of a Single Fiber

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ABSTRACT: Small-angle light scattering (SALS) was shown to be a suitable analytical method for characterizing submicrogram-sized samples of single textile fibers. Fibers from a variety of different generic groups were examined by SALS and produced widely differing scattering patterns. These differences were of such magnitude as to allow discrimination between all fibers examined.

KEY WORDS: criminalistics, fibers, lasers

Scattering methods using various forms of electromagnetic radiation are often essential to the elucidation of material structure. The wavelength of the radiation chosen for a particular study must be approximately the same size as that of the structural unit to be examined. For example, X-rays of wavelength about 0.1 nm are used in X-ray diffraction for study of structures ranging in size from a few thousandths to a few tenths of a nanometre. On the other hand, visible light of wavelength 400 to 700 nm is used in small-angle light scattering (SALS) to investigate structures ranging in size from about 10^2 nm (0.1 μm) to about 10^5 nm (100 μm). This range includes both internal morphological structures, surface structures, and fibers in toto. While all of these objects are within the observation range of optical and scanning electron microscopy, SALS offers the advantage of recording all accessible structural information within a single scattering envelope. As a result, the spatial intensity distribution of scattered light in SALS is a structural average for the whole sample and greater sensitivity may be achieved than with scanning electron microscopy.

Since the size range accessible to SALS includes the various morphological structures characteristic of polymers, SALS is widely used in structural studies of polymeric films [*1*]. In particular, the technique has proved useful in the characterization of spherulite morphology in films. For example, the average size of undeformed spherulites, the extension ratio of deformed spherulites, and the optical sign of spherulites are rapidly accessible by SALS [*1*].

The demonstrated success of SALS in the structural characterization of polymeric films led us to an investigation of the suitability of SALS for the characterization of single fibers

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in forensic science applications. The technique seemed especially attractive because it is inexpensive, rapid, and completely nondestructive. In addition, a single fiber a few millimetres in length (microgram and submicrogram mass) constitutes a suitable sample size for SALS.

In spite of the success of SALS in the structural analysis of polymer films, there have been few applications to the analysis of fiber structure. Some use of SALS on fibers has, however, been reported [*2–5*].

Here we report on the suitability of SALS for characterizing small lengths of single fibers. An introduction to the SALS method is included, as is an illustration of its use in examining fibers from a variety of different generic groups. Reports soon to follow will illustrate the use of SALS in the discrimination of fibers within a single generic group.

Apparatus and Experimental Procedure

The apparatus necessary for measurements of fibers by SALS is simple and inexpensive to construct. An apparatus suitable for most measurements may be constructed for well under $1000, and many components will already be available in most laboratories. A simple system involving photographic detection is illustrated in Fig. 1. A continuous-wave helium-neon laser is used as the high-intensity light source. A low-power laser suffices, and 2 to 4 mW is adequate power. The laser offers a highly monochromatic light source of 632.8-nm wavelength. If the laser used does not emit polarized light, a polarized filter must be placed between the laser and the sample normal to the beam in order to plane-polarize the laser light. The fiber must be mounted in a holder so that it can be rotated at various angles to the incident polarized radiation. A washer with a 5- to 10-mm-diameter hole acts as a convenient holder and provides an opening larger than the diameter of the incident laser beam, which is usually 0.75 to 1.5 mm.

The fiber should be mounted on the detector side of the ring to minimize forward scattering from the washer. The fiber should be secured at the top with a clip, tape, or other means and then allowed to hang vertically with a small weight (about 0.1 g) clipped or taped to the lower end to allow relief of torsion in the fiber by untwisting. The fiber may be photographed either hanging or attached to the holder at the bottom for extra stability. A major advantage in allowing the fiber to hang vertically with a small weight on its end is in the comparison of crimped fibers, which requires the extension of crimp for each sample in a reproducible manner. By maintaining constant length for each fiber sample and by adding to each an identical load, the degree of crimp extension can be reproduced well within the limits required for forensic science analysis.

Fiber movement must be completely eliminated, so it is usually necessary to work in an unvented room and shield the sides and top of the sample holder with cardboard or some other material for added protection from air movement. A polarizer that functions as an "analyzer" is placed in the optical line between the fiber and the detector. It should have precise means of rotation so that its polarization direction may be varied 360 deg. The photographic detection system consists of a 100- by 125-mm (4- by 5-in.) Polaroid

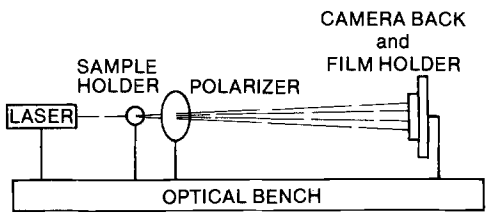

FIG. 1—*The SALS apparatus.*

film holder and camera back. In addition, for exposures of less than 1 s, it may be necessary to add a lensless shutter. In our laboratory, Type 55 P/N Polaroid film is used for all exposures. All components of this system are optically aligned by mounting each on an optical bench 1 to 2 m (3 to 6 ft) long. It is necessary to mount the laser on a stage with adjustable lateral translation so that the beam may be critically centered on the fiber sample. Sample-to-film distance may be easily varied to provide patterns at various angles of scattering. These optical components may be purchased from a variety of sources.[2]

A photographic image composed of many reflections is obtained in SALS of fibers and provides much information about the sample. The location of reflections is designated in a way identical to that of other electromagnetic scattering techniques. That is, three general reference areas are identified—the transmitted primary beam, meridian, and equator—and two angular displacements from these points are specified, as illustrated in Fig. 2. The radial angle θ designates angular displacement of the reflection from the center of the transmitted beam, and the azimuthal angle ϕ designates angular displacement of the reflection from one of the other reference points, usually the meridian. The radial angle θ may be calculated by the relationship

$$\tan \theta = d/D$$

where d denotes the distance of the reflection from the center of the primary transmitted beam to the reflection and D denotes the sample-to-film distance. The azimuthal angle ϕ may be measured directly with a protractor.

While both the sample and analyzer may be rotated continuously through 360 deg, most experiments require alignment in only two discrete arrangements. The fiber axis is held vertical in both arrangements and is parallel to the polarization direction of the incident radiation. For one arrangement, the polarization direction of the analyzer is set vertical, and for the other it is set horizontal. These two arrangements are referred to as Vv and Hv, respectively, where the large letter refers to the polarization direction of the analyzer and the small letter refers to that of the incident beam. There are occasions, however, when other alignments of sample and analyzer are necessary.

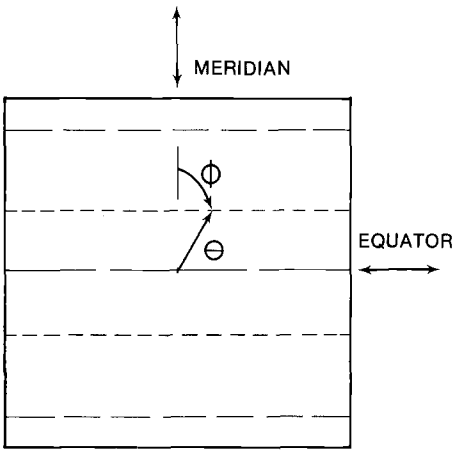

FIG. 2—*Radial (θ) and azimuthal (ϕ) angle specifications of a scattering maxima.*

[2] For example, the Newport Research Corp., Fountain Valley, Calif. and The Ealing Corp., South Natick, Mass.

To record a pattern, sample and polarizer alignments are selected and then the room is darkened completely. The incident laser beam is centered on the fiber sample, the sample-to-film distance selected, the film loaded, and an exposure made. Although the incident beam diameter is many times larger than that of the fiber, and the beam completely bathes the fiber, the beam must be carefully centered on the fiber to obtain a reproducible pattern. Rough alignment can be accomplished by moving the laser position laterally until the scattering intensity is maximized. Fine alignment may then be made by adjusting the laser position until interference fringes, if present, are symmetrical about the transmitted beam. In many cases, it is necessary to make an exposure to observe the interference fringes, as they are often very light. Figures 3 and 4 show the use of this method.

Data Analysis

Figures 3 through 11 show Hv patterns of 1-cm lengths of single fibers from various generic groups mounted in air with the fiber axis vertical. One can readily appreciate

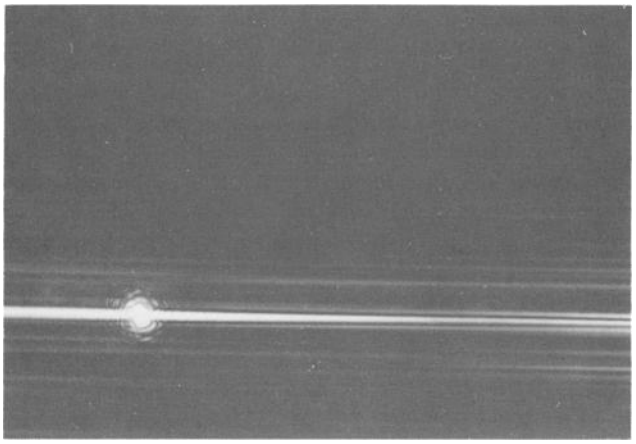

FIG. 3—*The Hv pattern of an acrylic fiber suspended in air (fiber axis vertical).*

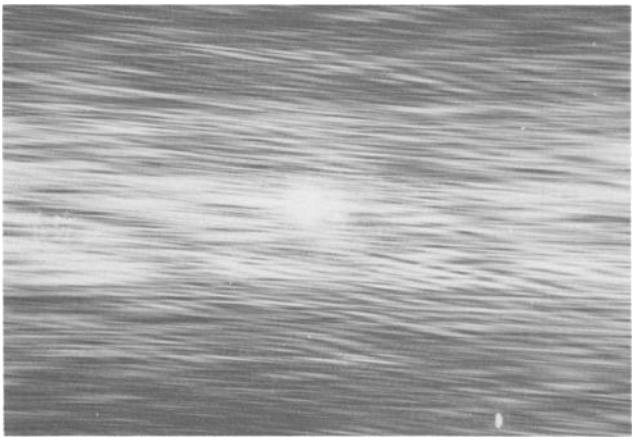

FIG. 4—*The Hv pattern of a cotton fiber suspended in air (fiber axis vertical).*

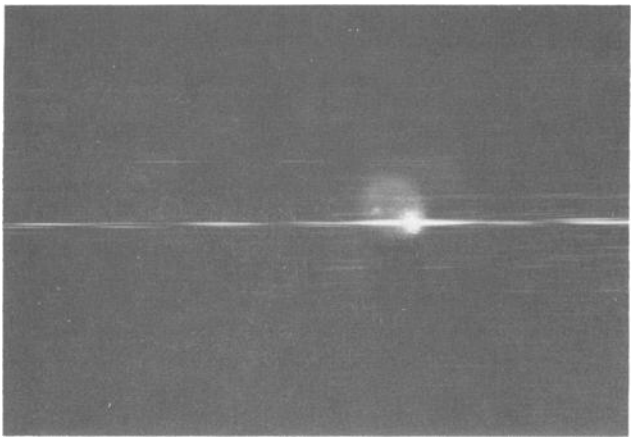

FIG. 5—*The Hv pattern of a glass fiber suspended in air (fiber axis vertical).*

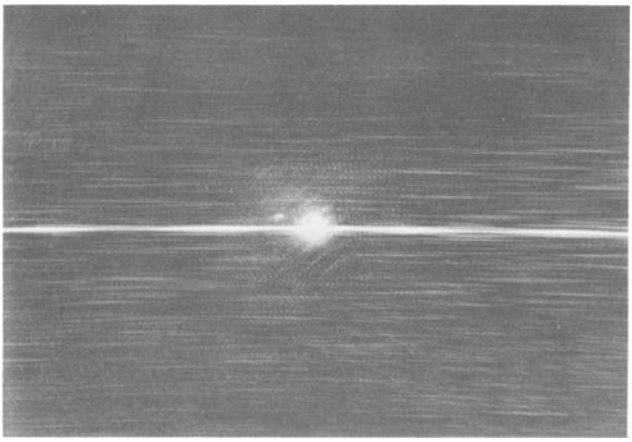

FIG. 6—*The Hv pattern of a polyester fiber suspended in air (fiber axis vertical).*

the sensitivity of SALS to fiber differences. General features of the photographs to be noted are the overall amount of scatter, discontinuity of lines (line splitting), azimuthal tilt of reflections, and overall symmetry of the scattering patterns. In addition, many photographs are characterized by unwanted scatter resulting from stray reflections. For example, the white spot in the lower right-hand corner of Fig. 4 is a result of a light reflection from the apparatus. Unwanted scatter may also arise from the sample holder. The halo at very low angles in Figs. 5, 7, and 8 results from this cause.

The overall amount of scatter varies greatly among various fibers, ranging from a small amount for the glass fiber of Fig. 5 to the largest amount for wool (Fig. 9) and cotton (Fig. 4). The amount of scatter from any substance is a function of the total number and magnitude of fluctuations in density or anisotropy. Consequently, the scatter in these figures can be interpreted as resulting from a small number of fluctuations or fluctuations of very small magnitude, or both, in the case of the glass fiber, and from a large number of fluctuations or fluctuations of large magnitude, or both, in the case of the cotton and wool fibers.

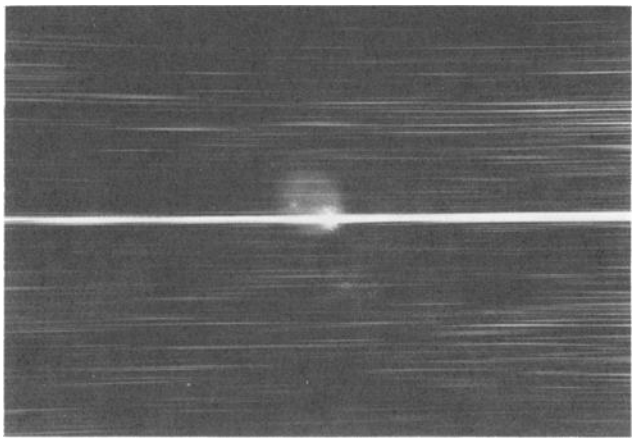

FIG. 7—*The Hv pattern of an aramid fiber suspended in air (fiber axis vertical).*

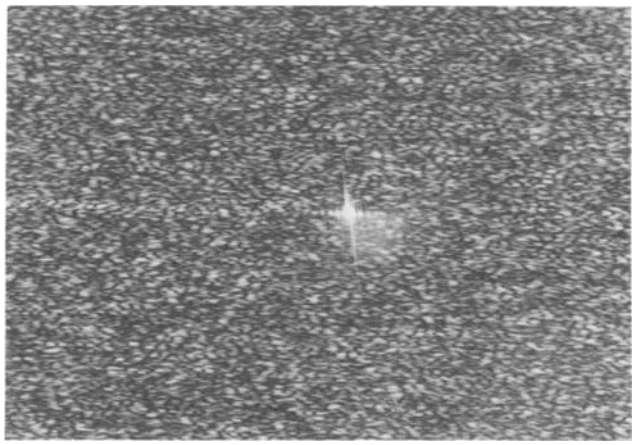

FIG. 8—*The Hv pattern of a saran fiber suspended in air (fiber axis vertical).*

Line continuity also varies greatly among the fibers. The acrylic fiber of Fig. 3 shows strong continuity at low angles while the lines increase slightly in discontinuity at higher angles. On the other hand, the saran fiber of Fig. 8 shows extreme discontinuity that is nearly uniform throughout the entire angular range observed. Line splitting has been ascribed to irregularity in scattering objects [*2*]. That is, deviation of fluctuations from the mean value is great. The other fibers shown are intermediate between the acrylic and saran fibers in line splitting. When the SALS pattern is characterized by severe splitting, it becomes difficult to determine the line segments that correspond to a single layer line. If a layer line can be identified, however, line splitting can be used to characterize the fiber. Standard mathematical methods in electromagnetic scattering may be used to calculate the size of the object causing the layer line splitting once the radial location of the reflection, θ, is known. That is,

$$b = n\lambda/\sin\theta$$

FIG. 9—*The Hv pattern of wool fiber suspended in air (fiber axis vertical).*

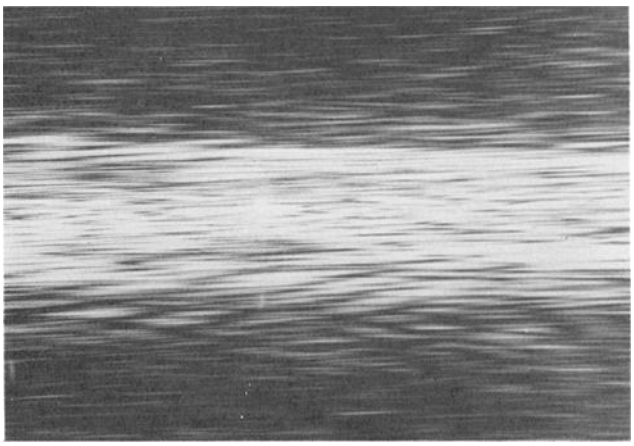

FIG. 10—*The Hv pattern of an acetate fiber suspended in air (fiber axis vertical).*

where

b = size of the reflecting entity,
$n = 1, 2, 3, \ldots,$
λ = wavelength of light used, and
θ = radial location of the reflection.

As an example illustrating the use of this equation, the equatorial reflection of Fig. 5 is used to calculate the size of the entity causing the reflection as follows: for $D = 300$ mm and $d = 12.0$ mm, $\theta = 2.29$ deg and since the reflection measured was the first minimum, $n = 1$, and it is found that $b = 15.8$ μm. This distance corresponds precisely to the diameter of the fiber. Hence the equatorial reflection was produced by scattering from the fiber in toto. Other reflections may be treated similarly.

Some SALS patterns are characterized by reflections having azimuthal tilt. Figure 4, a pattern from a cotton fiber, demonstrates much line tilt. This can be explained by a

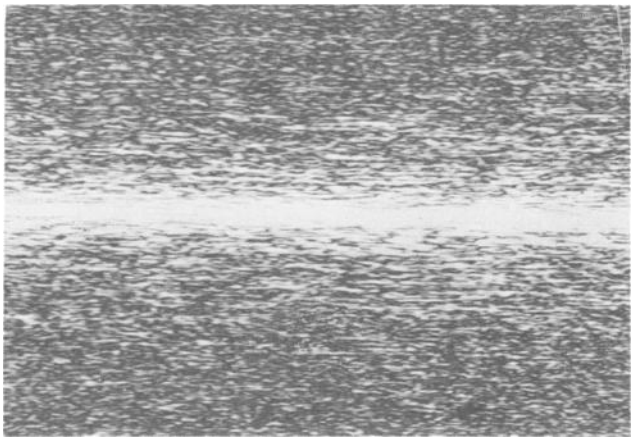

FIG. 11—*The Hv pattern of a polypropylene fiber suspended in air (fiber axis vertical).*

regular tilt of fibrils within the fiber caused by the characteristic twisted, convoluted structure of cotton.

An examination of the overall scattering pattern sometimes reveals asymmetry, as illustrated in Fig. 9, a pattern from a wool fiber. The unidirectional nature of scales on the wool fiber surface produces a pattern with a greater amount of scatter on one half of the picture than on the other half.

Total scattering resulting from a fiber is a function of surface scatter, cross-sectional shape, and internal structure within the size limitations imposed by the radiation used. Scatter from the surface of the fiber may be effectively blocked out by immersing the fiber in oil of the same refractive index as the fiber between two microscope cover slides. A set of Cargille refractive index liquids provides a convenient source of liquids for this use. The fiber surface is rendered "invisible" to the laser light, in the same manner that a fiber is rendered more visible during optical microscopy by a mountant of widely differing refractive index. Figure 12 is an Hv pattern from a polypropylene fiber mounted in an oil of refractive index nearly equal to that of the fiber. In comparing Fig. 11 to Fig. 12, one can readily determine that portion of the pattern resulting from surface scatter.

In general, scattering of light by a material results from fluctuations in sample density and anisotropy [*6*]. While density fluctuations provide information concerning sample homogeneity, anisotropy fluctuations provide information concerning the shape and orientation of anisotropic scattering units. The Vv scattering mode allows observation of scatter resulting from both density and anisotropy fluctuations while the Hv mode allows observation of scatter resulting from only anisotropy fluctuations. Thus scatter caused by density fluctuations may be blocked out experimentally. Figure 13 shows a Vv pattern from a polypropylene fiber mounted in air. Comparing Fig. 13 with Fig. 11 shows that the general character of scattering is changed. In addition, scatter resulting from density fluctuations can be seen at very low radial angles. It is interesting to note that this scatter is found around the primary beam on both sides of, but not on, the meridian.

Summary

Small-angle light scattering is a physical method of analysis suitable for analysis of a single fiber. It is completely nondestructive, inexpensive, quick, and simple. The work

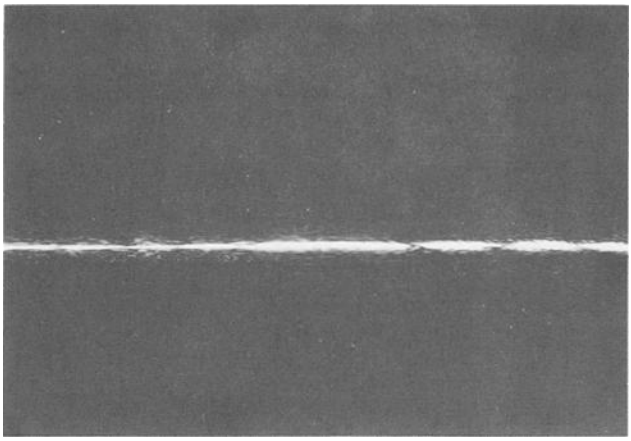

FIG. 12—*The Hv pattern of a polypropylene fiber mounted in oil (fiber axis vertical).*

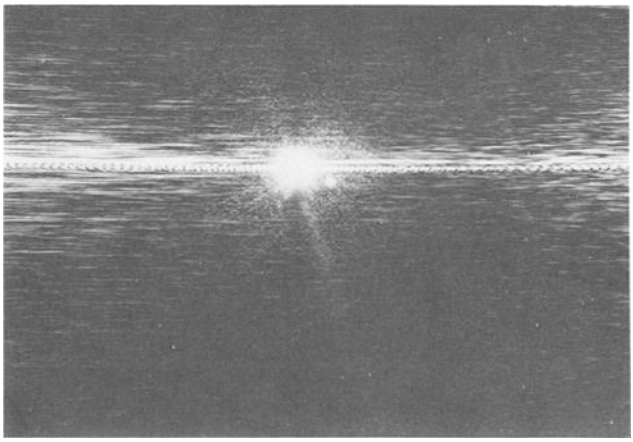

FIG. 13—*The Vv pattern of a polypropylene fiber suspended in air (fiber axis vertical).*

summarized in this paper has demonstrated the utility of SALS for characterization of fibers from different generic groups.

Present work in our laboratory is involved in demonstrating the power of SALS to discriminate between fibers within a single generic group. In addition, work is being carried out to demonstrate the power of SALS to discriminate between fibers of identical commercial origin subjected to differing laundering practices and outdoor exposure. Reports of this work will follow very soon.

Acknowledgments

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