SPECKLE PATTERNS IN SMALL ANGLE LIGHT SCATTERING: THE SPATIAL AUTOCORRELATION FUNCTION

Jaroslav HOLOUBEK, Josef BALDRIAN

Institute of Macromolecular Chemistry, Czechoslovak Academy of Sciences, 162 06 Prague 6

Received June 20th, 1985

Dedicated to Dr B. Sedláček on the occasion of his 60th birthday.

The study deals with the determination of the spatial autocorrelation function of speckle patterns caused by the small-angle light scattering from polymer films. The autocorrelation function determines the shape, size and anisometry of the speckle. The effect of the inner structure and orientation of samples (polypropylene foil, poly(decamethylene terephthalate) and a sample of polypropylene filaments) is discussed; it is shown that under the usual experimental conditions the spatial autocorrelation function of speckle patterns can be determined on the basis of the van Cittert–Zernike theorem of the classical coherence theory. The good agreement between the theoretical and experimental dependences of anisometry, the angular dependence of speckle size and the dependence of speckle size on the sample thickness confirm the suitability of a uniform description based on the classical theory of coherence. From the standpoint of the theory of speckle effect, the results presented in this study allow us to infer that in the light scattering from polymer films under usual conditions the assumptions of the application of the central limit theorem are fulfilled: in the scattering volume there is a sufficient number of scattering units, and path fluctuations due to the scattering foil exceed the wavelength of light.

The scattering of coherent light from an inhomogeneous medium gives rise to speckle patterns¹ (SP) which are a result of the interference of partial waves scattered from various regions of the inhomogeneous medium. The methods in which this phenomenon is utilized can be generally characterized as techniques based on measurements of the intensity correlation of scattered light, such as holographic interferometry, speckle interferometry, speckle photography, laser Doppler velocimetry or correlation spectroscopy. While the former three techniques predominantly analyze spatial aspects of intensity changes, the latter two deal with the time or frequency dependences of the intensity fluctuations of light scattered from an inhomogeneous medium. Although each separate technique has been successfully applied, their mutual relationship had not been investigated until very recently. Lately, the problem was being solved by means of a uniform description in terms of the classical theory of coherence^{2,3}. In the study of the dynamics of polymer systems correlation spectroscopy (the quasi-elastic light scattering technique) is particularly used⁴. The spatial analysis of SP was

Collection Czechoslovak Chem. Commun. [Vol. 50] [1985]

applied to polymer systems in connection with the investigation of deformation⁵, and also with dynamic processes with long correlation times^{6,7}.

In this study we examine the statistical properties of stationary SP caused by the light scattering from solid polymers as observed in the small-angle light scattering and demonstrate their basic properties. It is shown that under the assumptions given below the spatial autocorrelation function of SP may be determined by means of the van Cittert–Zernike theorem known from the classical coherence theory⁸. The autocorrelation function for SP of the H_v type (the scattering object is placed between crossed polarizers) is also determined, and problems related to the determination of the autocorrelation function for similar systems are discussed⁹. It is also shown that under the usual experimental conditions the statistical properties of SP due to the small-angle light scattering are in a good agreement with the assumed properties derived from the random phase screen theory^{10,11}.

THEORETICAL

In this chapter we discuss some second-order statistical properties of speckle which characterize the coarseness of the spatial speckle structure. We restrict ourselves to SP caused by the scattering of coherent light from objects which have the character of a deep random phase screen, *i.e.* the scattering object gives rise to path fluctuations exceeding the wavelength of light (the so-called fully developed speckle patterns in the statistical model given by Goodman)¹. Regardless of the fact whether SP arise by free-space propagation of scattered light or by imaging, the amplitude of the electric field A(x, y, z) consists of a large number of contributions from various scattering regions of the object with a random phase. We therefore search for the eventual complex amplitude A(x, y, z) at the observation point x, y, z as the sum of many elementary contributions N with the amplitude $a_k/(N)^{1/2}$ and phase Φ_k , assuming that (a) the amplitude and phase of the k-th elementary contribution are mutually independent and also independent of the amplitudes of phases of all other elementary contributions, (b) the phases Φ_k are uniformly distributed on the primary interval $(-\pi, \pi)$.

The problem of a complex addition of many elementary contributions which form the resulting field A is identical with the classical problem of a random walk in the plane. If the number of elementary contributions is sufficiently high $(N \to \infty)$, the central limit theorem may be used, with the result that the resulting field A has the Gaussian probability density (the amplitude A of the scattered field is a circular complex Gaussian random variable). The corresponding statistical properties of the intensity in a polarized SP are determined by employing the known procedure of transformation of random variables. The same model is the basis of the classical coherence theory⁸, if the term "scattering object" is replaced with the term "extended, quasi-monochromatic, thermal light source". From this equivalence it follows³ that field correlations of such SP are equivalent to the mutual intensities in radiation from quasi-monochromatic, thermal light source. Hence, field correlations (and thus also the autocorrelation functions of SP intensity) may be determined from the mean intensity distribution over the source by using the van Cittert–Zernike theorem^{8,10-12}.

Let r_1 , r_2 denote the distance from the point (ξ, η) in the plane \sum (Fig. 1a) to the points P_1 and P_2 ; for the optical field produced by an extended incoherent quasi-monochromatic source we have, then,

$$y_{12}(P_1, P_2) = C / [I(P_1) I(P_2)]^{1/2} \int_{\Sigma} I(\xi, \eta) \exp [ik(r_1 - r_2)/r_1 r_2] d\xi d\eta , \qquad (1)$$

where γ_{12} is the complex degree of coherence at the points P_1 , P_2 , $I(P_i)$ are the intensities at the points P_i . Relation (1) means that the complex degree of coherence (normalized autocorrelation function of the field in the terminology of the speckle theory) equals the normalized complex amplitude in the diffraction experiment, when the source is replaced with a diffraction aperture identical by its size and shape with the source; the amplitude distribution in the wave front is proportional to the intensity distribution across the source.

If the dimensions of the source and the distance between P_1 and P_2 are small compared with the distance between the source and the plane containing the points P_1 and P_2 , the degree of coherence is a normalized Fourier transform of the intensity function of the source. The explicit relation for a uniform rectangular incoherent source, of width 2a and length 2b, is¹²

$$\gamma_{12}(P_1, P_2) = \exp(ik\beta) \operatorname{sinc} \left[ka(x_1 - x_2)/r \right] \operatorname{sinc} \left[kb(y_1 - y_2)/r \right], \qquad (2)$$



FIG. 1



where the notations sinc $x = \sin x/x$ and $\beta = [(x_1^2 + y_1^2) - (x_2^2 + y_2^2)]/2r$ are used. Similarly, for a circular incoherent source of the radius ρ we have¹²

$$\gamma_{12}(P_1, P_2) = \exp(ik\beta) 2J_1(k\alpha\varrho/r)/(k\alpha\varrho/r), \qquad (3)$$

where $\alpha = [(x_1 - x_2)^2 + (y_1 - y_2)^2]^{1/2}$, J_1 is the Bessel function of the first kind. Usually, the mean speckle size is regarded as the value of the argument at which the functions sinc² $(ka \Delta x/r)$ or sinc² $(kb \Delta y/r)$ (relation 2), or the function $J_1^2(k\alpha q/r)$ (relation 3) fall to zero for the first time. It should be noted that relation (2) predicts the anisometry of the speckle shape, *i.e.* a different dimension in the direction of the y and x axes.

So far, we have considered an extended incoherent source of radiation. For a threedimensional incoherent source the solution to this problem can be found by using the so-called Projection Theorem of the Fourier transform¹². The Pojection Theorem states that the two-dimensional Fourier transform of a projected structure is the central section through the three-dimensional transform which is perpendicular to the direction of view. For a three-dimensional chaotic source shaped as a rectangular box with the sides L_{ξ} , L_{η} and L_{ϱ} (Fig. 1b) the size of the coherence area A_c (area where $\gamma_{12} > 0$) at the distance $r \gg L_{\xi}$, L_{η} , L_{ϱ} is given as¹³

$$A_{c} = \Omega_{coh}r^{2} = (r^{2}\lambda^{2})/(L_{\xi}L_{\varrho}\sin\Theta\cos\Phi + L_{\xi}L_{\varrho}\sin\Theta\sin\Phi + L_{\xi}L_{\eta}\cos\Theta), \qquad (4)$$

where $\Omega_{\rm coh}$ is called the coherence solid angle, Θ is the scattering angle and Φ is the angle between the direction of view and the axis η . In the scattering plane xz, $\Phi = 0$, and it may be written

$$A_{c} = (\lambda r)^{2} / (L_{\xi} L_{\varrho} \sin \Theta + L_{\xi} L_{\eta} \cos \Theta) =$$

= $(\lambda r / L_{\xi}) (\lambda r) / (L_{\eta} \cos \Theta + L_{\varrho} \sin \Theta) = A_{\perp} A_{\parallel},$ (5)

where $A_{\perp} = \lambda r / L_{\xi}$ and $A_{\parallel} = \lambda r / (L_{\eta} \cos \Theta + L_{\varrho} \sin \Theta)$, A_{\perp} , A_{\parallel} denote the speckle dimensions perpendicular and parallel to the scattering plane.

Although the explicit relations for the complex degree of coherence $\gamma_{12}(P_1, P_2)$, and thus also for the spatial autocorrelation function are comparatively simple, an exact determination of this function from experimental data is not simple⁹. Because of the statistical nature of SP, it is necessary to process a large amount of data in order to obtain an adequate estimate of these functions. The so far most complete measurement of second-order statistical quantities (autocorrelation function and power spectrum) has been made by McKechnie¹⁴. His study has two main drawbacks as regards our own purpose. It is limited to the statistically stationary speckle, when statistics pertaining to one point in the pattern are identical to those at any other point; in other words, ensemble averages of any quantity can be obtained directly from its space averages by scanning across a single SP.

For the SALS SP, we may expect a slightly nonstationary character (dependence of the autocorrelation function of SP, and hence of the speckle size on the scattering angle in the sense of relation (4)). It will be shown that in many cases this nonstationary character is weak, and the procedure just described (space averaging) leads to good estimates of statistical quantities. Another problem yet unsolved consists in the fact that SALS SP has a variable nonzero average value, which affects the shape of the autocorrelation function, as also is shown below.

EXPERIMENTAL

SP formed by the small-angle light scattering from three types of samples were analyzed. Two types of samples were foils, one of isotactic polypropylene 160 μ m thick, another of poly(decamethylene terephthalate), about 15 μ m thick, with circular spherulites¹⁵; the third sample was made from a bundle of polypropylene filaments 30 μ m in diameter, wound in parallel to each other and placed in an immersion with the refractive index n_D 1.514. The experimental arrangement used allowed us to influence the shape of the illuminating beam. The collimated laser beam passed through slits of various shape and size. The slits used were a circular one, 2.0 mm in diameter, a square one with the side 1.1 mm, and rectangular slits 5.5 . 1.1 mm and 5.5 . 0.6 mm. The light source was a He–Ne laser Spectra Physics 124 A. We used a lens L1 with the focal distance, f, 50 mm and L2 with f 210 mm. The sample — detection plane distance was 160 mm. The speckle patterns were recorded with a Polaroid 665 P/N film.

In order to determine the autocorrelation function of the intensity fluctuations of scattered light, a photograph of the SP of spherulitic structure of isotactic polypropylene was analyzed for a sample illuminated with circular beam. A scattering pattern of the H_v type was measured (the vertical linearly polarized incident light, scattered light with the horizontal plane of polarization). We chose direct determination of the spatial autocorrelation function of intensity fluctuations by means of calculation^{14,16}

$$|\gamma_{12}|^2 = 1/(N-j)\sum_{i=1}^{N-j} \Delta I_i \,\Delta I_{i+j}, \quad j = 0, 1, 2, ..., n.$$
 (6)

The *n* value was chosen by using the sampling theorem (Nyquist formula). The photographic record was scanned with a digital microdensitometer using a long narrow slit of the length *L*. It was necessary to choose (a) the sampling interval, *d*, (b) the total number of data, (c) the scanning-slit dimensions. The negative was scanned with a digital automatic photometer AD-1 (SYNTEX, USA), the data were recorded on a magnetic tape and then computations were carried out. Using an approximate speckle size from the geometry of the experiment and from the determination of size values with an optical microscope, the average dimension $\langle A_c \rangle$ was found to be 70 µm. The following parameters of the experiment were therefore chosen: *a*) sampling interval 32 µm, *b*) a total number of data about 10³, *c*) slit width 32 µm, slit length 1.5 mm. Such choice of the estimate of the autocorrelation function as a function of the total sampling length and of the effective bandwidth).

We did not attempt to raise the accuracy of determination of $|\gamma_{12}|^2$ by processing more measurements, because it appeared that the dependence of $|\gamma_{12}|^2$ is greatly affected by the choice

of the average intensity value which varies considerably in the measured angular range (Fig. 2)*. During sampling, a correction was made which respected the fact that the intensity values were scanned from a planar record, *i.e.* the sampling interval was modified depending on the scattering angle, because the varying distance sample-position in the detection plane affects the dimensions of the speckle (Fig. 3).

RESULTS AND DISCUSSION

The dependence of the shape of $|\gamma_{12}|^2$ on the average intensity value is shown in Fig. 4. The curves gradually represent $|\gamma_{12}|^2$ obtained from the deviations $\Delta I_j = I_j - \langle I \rangle$, where $\langle I \rangle$ is taken as the mean from (a) 1 016, (b) 400, (c) 200, (d) 100 and (e) 20 values from the vicinity of the given sampled point; each time, one half of the values precedes the given point j and one half follows it. It can be seen that the choice of $\langle I \rangle$ has a considerable influence on the shape of $|\gamma_{12}|^2$. The following Fig. 5 shows the





Scheme of an experiment used in the investigation of the effect of shape of the illuminating beam on the autocorrelation function of intensity fluctuations: 1 laser, L1 lens, 2 pinhole, L2 projection lens forming the parallel beam, 3 aperture determining the form of the illuminating beam, 4 sample, 5 detection plane, 6 wide-angle detection by circular camera

Fig. 4

Dependence of the shape of the autocorrelation function on the average intensity value $\langle I \rangle$; numbers are given of the sampling points used in the determination of the mean value (a) 1016, (b) 400, (c) 200, (d) 100, (e) 20

* See insert cn the page 2938 at the end of this issue.

2878

shape of $|\gamma_{12}|^2$ for a generalization of the two-dimensional random checkerboard process¹⁷ used as a suitable model for a variety of image processing applications. The model is characterized by two key parameters λ and ν which are graphically related to the image properties. While λ characterizes the size of the checkerboard square, the parameter ν given in Fig. 5 indicates the degree of abruptness across an edge boundary. For large and negative ϱ there exists an almost black-and-white or white-and-black transition between the adjacent squares of the checkerboard. This fits in with theoretical views about the unity contrast of fully developed SP when intensity fluctuations are equal to the mean value. Simultaneously, it follows that, when comparing arbitrary autocorrelation function, one should use an identical way of determination of the mean intensity value if the comparison is to be feasible.

Although we obtained a qualitative agreement with the assumed results (the autocorrelation function of scattered light intensity behaves similarly to the autocorrelation function of the generalized two-dimensional random checkerboard process), the results of determination of the autocorrelation function are not quite satisfactory. The results presented in this study do not allow us to draw well-founded conclusions about the relation between the shape of the autocorrelation function and structural details of the sample. Fig. 4 also indicates difficulties involved in the determination of speckle dimensions from zero value of the autocorrelation function, as this value greatly depends on the value of $\langle I \rangle$.

We therefore carried out experiments which graphically demonstrated properties of the autocorrelation function by using two different types of samples which allowed us to investigate parameters of the autocorrelation function under rather extreme

FIG. 5

Dependence of the shape of the autocorrelation function of a random two-dimensional checkerboard process on t; the parameter, ρ , characterizing the degree of abruptness across the edge boundary¹⁷. Normalized displacement, t, is expressed in the checker board square edge units $\begin{array}{c}
10 \\
p - 0 9 \\
1 \\
2 \\
3 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
- 0 \\
-$

conditions. a) To estimate the dependence of the autocorrelation function on structural details of the sample, we used the poly(decamethylene terephthalate) polymer foil with ringed spherulites which contains scattering patterns with three orders of diffraction maxima corresponding to the respective structural morphological units¹⁵. The results of the investigation of this sample are shown in Fig. 6*. The magnified detail on the left-hand side of the figure shows the speckle shape obtained with the given shapes of the irradiating beam (visible also in the centre of SP). In the cases 6a, b an identical rectangular aperture with different orientation was used (rotation by 90°). The results confirm that the autocorrelation function is quite unambiguously connected with the shape of the illuminating beam in the sense of the van Cittert--Zernike theorem, *i.e.* the autocorrelation function is proportional by its form, extent and orientation to the intensity profile of the illuminating beam (it is its diffraction pattern). As shown in Fig. 6, the shape and size of the speckle are the same within the whole angular range observed and are unaffected by structural details discernible by the analysis of the individual diffraction orders. From Fig. 6 it follows that the scattering object acts as an incoherent source in the sense that light scattered from various "points" of the object has uncorrelated random phases. By a "point" we mean here a region of a few wavelengths in extent corresponding morphologically to an individual ringed spherulite. b) Another effect which might influence the autocorrelation function is the pronounced anisotropy of the scattering system. This effect was investigated for a sample prepared from a layer of polypropylene monofilaments wound parallel to each other and placed in an immersion with the refractive index n = 1.516, in order to stress the effect of the inner structure of the filaments and suppress effects of scattering from the filament surface. Fig. 7a, b^{**} illustrates the effect of orientation of the sample if the illuminating beam is rectangular (vertically oriented slit with dimensions a) 5.5.1.1 mm, b) 5.5.0.6 mm). In Fig. 7a the filaments are oriented in parallel with the longer side, in Fig. 7b they are oriented perpendicularly to the longer side of the rectangle. In both cases the speckle shape and size (and thus also the autocorrelation function of the intensity fluctuations of scattered light) correspond to the size and orientation of the illuminated region of the sample, irrespective of the orientation of the sample. It should be mentioned that the integral small-angle light scattering (radiation envelope with smoothed-out speckle) reflects orientation of the filaments, *i.e.* the dependence $I(\Theta, \varphi)$ for Fig. 7a corresponds to the dependence $I(\Theta, \varphi + \pi/2)$ in Fig. 7b, where Θ is the polar and φ is the azimutal scattering angle. Fig. 7c demonstrates SP in the illumination with a circular beam (beam diameter 2.0 mm) with horizontally oriented filaments, Fig. 7d shows the effect of the square aperture 1.1. 1.1 mm with vertically oriented filaments. Similarly to Figs a, b, the size and shape of the illuminating aperture play the decisive role.

* See insert on the page 2938.

^{**} See insert on the page 2938.

The experiments described in this study allow us to state that monochromatic illumination of polymer foils and filaments with a supermolecular structure with coherent light under the usual experimental conditions (nonfocussed laser beam, sample thickness from tens to hundreds of microns, small-angular region of scattered light) gives rise to a speckle pattern where the speckle shape and size (specified by the spatial autocorrelation function of the intensity fluctuations of scattered light) are determined by the shape, size and orientation of the aperture which restricts the illuminating beam, and thus in accordance with the van Cittert-Zernike theorem.

Up to now, we have regarded the scattering foil as sufficiently thin for the scattering volume to be considered a planar source. The photographs confirm that such assumption is realistic, because in the angular region studied no angular dependence of the speckle size can be observed. The dependence of the speckle size on the scattering angle calculated by means of relation (4) for a chaotic source having the form of a rectangular prism $(L_{\xi} = L_{\eta} = a, L_{o}$ is foil thickness) is shown in Fig. 8. The A_{\parallel} and A_{\perp} values respectively denote the speckle dimension in the scattering plane and perpendicularly to it. It can be deduced from Fig. 8 that under the conditions usual in the investigation of small-angle light scattering from polymer foils the dependence of the speckle size on the scattering angle is small (the relative change in speckle size in the angular range $0-20^{\circ}$ is below 4% in this case), and is macroscopically unobservable, in good agreement with Figs 6 and 7. A different situation arises in the case of a focussed or narrow beam. This case is illustrated by Fig. 9. The dependence of the speckle size on the scattering angle, sample thickness and the size of the illuminating beam suggests that for the focussed illuminating beam and major sample thicknesses one may expect a pronounced angular dependence of the speckle size. In such case the relative





Theoretical dependence of the speckle size A_{\parallel} on the scattering angle Θ for a chaotic quasiplanar source; $r = 100 \text{ mm}, L_{\xi} = L_{\eta} =$ $= 1.2 \text{ mm}, L_{o}$: 1 50 2 100 3 150 mm



change in the speckle size varies from 20 to 60% in the angular range $0-20^\circ$, *i.e.* is larger by an order of magnitude than in the case shown in Fig. 8. This effect has been proved indeed and is illustrated in Fig. 10*. A distinct dependence of the speckle size on the scattering angle can be seen with the naked eye, similarly to the anisometry of the speckle form (different A_{\parallel} and A_{\perp} values), especially in the angular range 20 to 50°. In the angular range $50-90^\circ$ the shape and size of the speckle do not correspond to a simple theoretical dependence (Fig. 9), probably due to deviations of the illuminated volume from an ideal cylindrical shape, and to the multiple light scattering. It should be mentioned that by neglecting the dependence of the speckle size on the sample thickness, one arrives at incorrect structural interpretations¹⁸.

The experiments reported in this study allow us to state that under the usual experimental conditions (monochromatic laser beam, foil thicknesses $20-200 \,\mu$ m, supermolecular structure up to tens μ m) SP arising in the small-angle light scattering from polymer foils possess spatial properties which may be derived by using the van Cittert-Zernike theorem known from the classical coherence theory. This means that the scattering volume acts as an incoherent source in the sense that light scattered from various points of the object has uncorrelated random phases. In such situation the spatial analysis of SP does not give direct structural information of the scattering medium (unlike the so-called non-Gaussian SP)¹⁹. However, also in these cases the deformation and dynamics of motion of the scattering object can be investigated by analyzing SP changes^{5,19}. From the standpoint of the theory of speckle phenomenon the results presented here confirm that fully developed SP can be obtained if there is a sufficient number of scattering sites in the illuminated volume which cause phase fluctuations larger than π and thus support the correctness of Pedersen's arguments²⁰ used in the discussion of the statis⁺ical theory of speckle patterns.

We are indebted to Dr I. Daniewska, Technical University, Warsaw, Poland, and Dr O. Durčová, Research Institute of Chemical Fibers, Svit, Czechoslovakia, who kindly supplied the samples.

REFERENCES

- 1. Dainty J. C., Ed.: Laser Speckle and Related Phenomena. Springer Verlag, Berlin 1975.
- 2. Erf R. K.: Speckle Metrology. Academic Press, New York 1978.
- 3. Pedersen H. M.: Opt. Acta 29, 105 (1982).
- 4. Chu B.: Laser Light Scattering. Academic Press, New York 1974.
- 5. Holoubek J., Sedláček B.: J. Polym. Sci., Polym. Phys. Ed. 18, 265 (1980).
- 6. Carroll P. J., Patterson G. D., Cullerton S. A.: J. Polym. Sci., Polym. Phys. Ed. 21, 1889 (1983).
- 7. Holoubek J.: Opt. Acta 31, 1283 (1984).
- 8. Born M., Wolf E.: Principles of Optics. Pergamon Press, Oxford 1964.
- 9. Moneva I., Michailov M.: Makromol. Chem., Rapid Commun. 2, 267 (1981).

* See insert on the page 2938.

- 10. Crosignani B., Di Porto P., Bertolotti M.: Statistical Properties of Scattered Light. Academic Press, New York 1975.
- 11. Asakura T., Takai N.: SPIE Vol. 243, 114 (1980).
- 12. Lipson H. S., Ed.: Optical Transforms. Academic Press, London 1972.
- 13. Cummins H. Z., Pike E. R., Eds.: Photon Correlation Spectroscopy and Velocimetry, p. 144. Plenum Press 1977.
- 14. McKechnie T. S.: Optik 39, 258 (1974).
- 15. Daniewska I., Hashimoto T., Nakai A.: Polym. J. 16, 49 (1984).
- 16. Dainty J. C., Shaw R.: Image Science. Academic Press, London 1974.
- 17. Modestino J. W., Fries R. W., Daut D. G.: J. Opt. Soc. Amer. 69, 897 (1979).
- 18. Voishvillo N. A., Shcherbakova N. I.: Opt. Spektrosk. 54, 185 (1983).
- 19. Holoubek J., Sedláček B.: Makromol. Chem. 185, 2021 (1984).
- 20. Pedersen H. M.: Opt. Acta 26, 149 (1979).

Translated by L. Kopecká.

Collection Czechoslovak Chem. Commun. [Vol. 50] [1985]









FIG. 6

Anisotropy of the speckle shape for a poly(decamethylene terephthalate) sample. On the left-hand side of the figure is a magnified detail: (a) circular aperture, (b) rectangular aperture, longer side of the rectangle is vertically oriented (cf. centre of the figure), (c) rectangular aperture, longer side horizontally oriented (cf. centre of the figure)

C



FIG. 7

Anisometry of the speckle shape for a sample made of polypropylene filaments: (a) circular aperture, filament axes horizontal, (b) square aperture, filament axes vertical, (c) rectangular aperture, longer axes oriented vertically, filament axes oriented vertically, (d) rectangular aperture, longer axis oriented vertically, filament axes oriented horizontally

a



b





d



FIG. 10

Experimental dependence of the speckle size A_{\parallel} on the scattering angle Θ for a chaotic threedimensional source. The polyethylene foil used, 380 µm thick, an illuminated volume with dimensions $L_{\xi} = L_{\eta} = 42 \ \mu\text{m}, \ L_{\rho} = 380 \ \mu\text{m}$. The photograph shows the angular regions (a) $(-17^{\circ}, 17^{\circ}), (b) (17^{\circ}, 51^{\circ}), (c) (51^{\circ}, 85^{\circ})$