# Spectral properties of speckle created by small-angle scattered light

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Use of a laser in small angle light scattering (SALS), results in scattering patterns which have a fine grain structure called 'speckling'. It appears that the existence of speckle is of a fundamental nature, and that complete removal of this phenomenon is not feasible without discontinuing use of laser or without a considerable reduction of the time or space coherence of the laser light[1]. However, many methods exist in which the properties of the medium are characterized from informatation obtained from the analysis of speckle[1–5]. It is known[6] that the properties of speckle produced by scattering on a rough surface allow us to determine the surface quality, i.e. the surface roughness. It therefore seems useful to employ the study of the decorrelation of speckle produced by two different wavelengths in order to examine the structural properties of polymer films. The increasing occurrence of inhomogeneities (inclusions, spherulite boundaries, microcracks, additives, etc.) should affect the distribution of phases of scattered light, and thus cause gradual decorrelation of speckle produced by scattered light which has passed through such a medium. Hence, there exists a possibility of quantitative testing of both the optical and mechanical 'quality' of the polymeric material, at least within a certain range of size and quantity of inhomogeneities. This report indicates that the first step has been taken in this direction; i.e. that two SALS speckle patterns created by two near-by wavelengths are quantitatively correlated. This fact, along with the recently observed correlated character of SALS speckle patterns in subsequent deformed states[3–5], provides for the possibility of a detailed investigation of processes which affect speckle.

To test the correlated character of speckle patterns, we used the method of pointwise filtering[3,4] of double-exposed SALS patterns at two wavelengths, $\lambda_1$ and $\lambda_2$ (see *Figure 1*). In the first exposure we recorded the scattering patterns $H_V$ of a film of isotactic polypropylene at $\lambda_1$, while in the second exposure the same plate was exposed at $\lambda_2$. The double-exposed speckle pattern thus obtained was processed by pointwise filtering, i.e. the laser light (wavelength $\lambda$) was allowed to pass through selected areas of the plate, and the Young's fringes in the far field were detected. The Young's fringes produced in this way, show that the two speckle patterns are correlated. The visibility of the fringe pattern depends both on the nature of the scatterer and the bandwidth of the light[6] and relates parameters of the fringe system to the structural properties of the film.

The following elements were used in the experiment: Ar laser (Spectra Physics 165) with wavelengths $\lambda_1 = 501.7$ nm, $\lambda_2 = 496.5$ nm tuned to the same power of 5 mW on these lines served as the source. The illuminated film was made of isotactic polypropylene, 160 $\mu$m thick, with an average size of spherulites $\sim 4$ $\mu$m. The double exposure was recorded on a Polaroid 665 P/N film. The negative of double exposure was processed by means of a He–Ne laser ($\lambda = 632.8$ nm) with power of $\sim 6$ mW. Light was allowed to pass through 15 areas in one quadrant of the scattering pattern of the $H_V$ type, situated in positions corresponding to the direction of the polarizer axis, of the analyser axis and at angles 30°, 45° and 60° to the polarizer axis. The results are shown in *Figure 2*. The relative displacement (given in %s), corresponding to the displacement of grains is related to the change of $\lambda_1$ to $\lambda_2$. The orientation of fringes given in degrees indicates a radial displacement (the direction of the displacement is perpendicular to the fringes, its magnitude is inversely proportional to the distance between them). The theoreti-

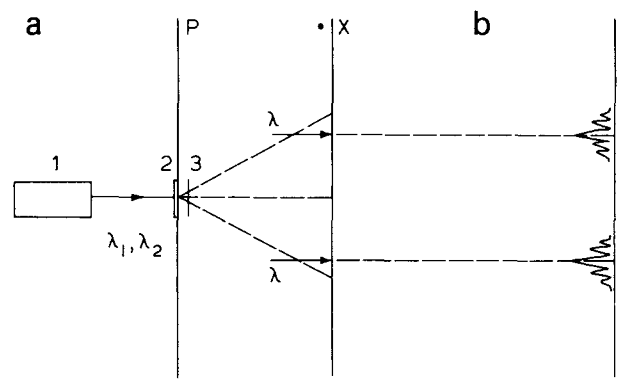


*Figure 1* Scheme of detection (a) and processing (b) of double-exposed speckle patterns. 1, laser; 2, sample; 3, analyser; P, object plane; X, plane of double exposed speckle patterns; Σ, recording plane of Young's fringes

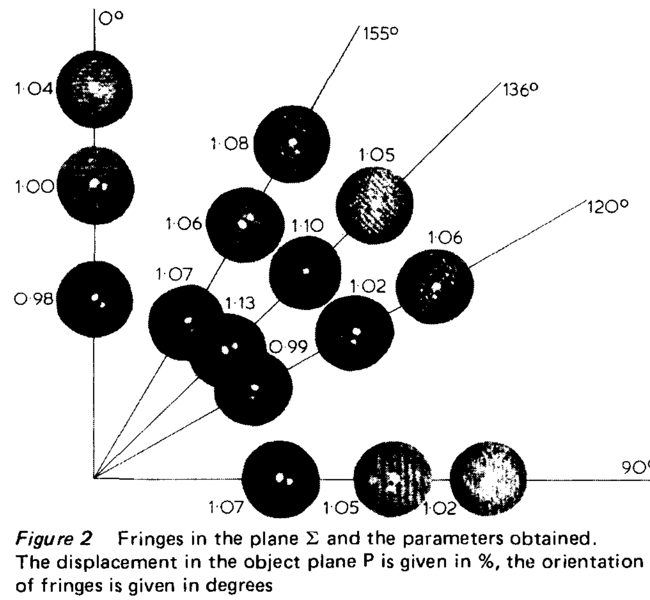


*Figure 2* Fringes in the plane Σ and the parameters obtained. The displacement in the object plane P is given in %, the orientation of fringes is given in degrees

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cal displacement related to the change of $\lambda_1$ and $\lambda_2$ is 1.05%, the average value obtained from the experiment is $(1.05 \pm 0.03)$%. There is good agreement between the experimental and theoretical displacements and orientation values.

The polychromatic illumination can be considered as made up of a discrete number of constituent wavelengths, each individually capable of producing a coherent speckle pattern. A reduced speckle, the one due to all wavelengths acting together, can be regarded as the incoherent addition of a number of coherent speckle patterns each displaced according to the individual wavelength, with the resulting effect of 'smoothing' of polychromatic speckle patterns[7].

The speckle produced by SALS may also be reduced by employing the method of time integration while the wavelength is fixed. Trivial yet important with respect to its consequences is the finding that in many cases such integration (analogous to the use of the random spatial phase modulator[8]) may be realized by motion of the scattering medium alone. This has been experimentally verified for light scattering from dispersions[9] (low-speed stirring of the sample). A similar procedure (rotation of the sample) may be employed also for unoriented solid polymers. The speckle noise in this case is reduced because of total averaging out of the interparticle interference function over the different particle configurations of the scattering units during the counting time or time of exposure.

The new findings reported here concern the spectral correlation of speckle created by SALS and point out the importance of investigation of the detailed properties of speckle with an aim to utilize the speckle analysis in the characterization of the medium by which it is created[3–5].

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# Solubility parameters of ternary solvent mixtures; calculation of the solvent composition with maximum polymer interaction


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Rigbi[1] has applied vector algebra to the calculation of the composition of binary or ternary solvent mixtures, leading to a maximum in the interaction with a given polymer in terms of the three-dimensional solubility parameter as proposed by Hansen[2] and applied by us to binary solvent mixtures[3]. The equations as given by Rigbi for each of the solubility parameter components in a ternary mixture appear not to be in accord with the requirement of symmetry with regard to the three component solvents (equation 8 in ref 1). Here, we give a more concise and mathematically more satisfying derivation of the optimum composition of ternary solvent mixtures.

As in Rigbi's paper the three-dimensional solubility parameters are represented by vectors; the three solvents are symbolized by $\underline{a}$, $\underline{b}$ and $\underline{c}$, the polymer by $\underline{p}$ and the optimum solvent mixture by $\underline{m}$. The end points of the respective vectors in the solubility parameter space are A, B, C, P and M. The requirement for maximum interaction is that PM is perpendicular to the plane through A, B and C; in other words PM $\perp$ AB and PM $\perp$ AC. In vector notation:

$$(\underline{m}-\underline{p})^{T}(\underline{b}-\underline{a})=0 \tag{1}$$

$$(\underline{m}-\underline{p})^{T}(\underline{c}-\underline{a})=0 \tag{2}$$

M lies in the ABC plane, from which follows that $(\underline{m}-\underline{a})$ is a linear combination of $(\underline{b}-\underline{a})$ and $(\underline{c}-\underline{a})$:

$$(\underline{m}-\underline{a})=\lambda_1(\underline{b}-\underline{a})+\lambda_2(\underline{c}-\underline{a}) \tag{3}$$

Elimination of $\underline{m}$ by substitution of equation (3) into equations (1) and (2) gives a set of two linear equations for $\lambda_1$ and $\lambda_2$:

$$\begin{aligned}(\underline{b}-\underline{a})^{T}(\underline{b}-\underline{a})\lambda_1+(\underline{c}-\underline{a})^{T}(\underline{b}-\underline{a})\lambda_2&=(\underline{p}-\underline{a})^{T}(\underline{b}-\underline{a})\\(\underline{b}-\underline{a})^{T}(\underline{c}-\underline{a})\lambda_1+(\underline{c}-\underline{a})^{T}(\underline{c}-\underline{a})\lambda_2&=(\underline{p}-\underline{a})^{T}(\underline{c}-\underline{a})\end{aligned} \tag{4}$$


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