Suppression of the speckle noise in solid polymer samples: a light scattering study

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Abstract. The speckle effect disturbs the measurement of spatial correlation functions in solid polymer samples by light scattering. To be able to extract the desired correlations from the measurements, the speckle noise must be suppressed. This is possible by moving the sample during the measurement. In this paper we demonstrate that a sufficient reduction of speckle contrast can be achieved even in samples of restricted dimensions or with a preferential direction.

Subject terms: light scattering; speckle suppression; sample translation; glassy polymers; semicrystalline polymers; oriented polymers.

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1 Introduction

Whenever a surface that is rough on the length scale of the optical wavelength is illuminated with coherent light, a granular pattern, usually called speckles, appears in the reflected light. This pattern is due to phase differences induced in the optical path by the random surface roughness. The random walk in the complex intensity plane induced by the statistically independent elementary scatterers leads to an exponentially decreasing intensity probability density function.[1] This means that the contrast of the speckle pattern is always unity, with the most probable intensity being zero.

In translucent solid samples, randomly distributed refractive index fluctuations lead to a light scattering that is superimposed by similar patterns. Almost all polymer samples (even glasses, which are usually thought of as being amorphous) contain inhomogeneities, which lead to optical path differences many times the wavelength of light and therefore show this speckle effect.

If one is interested in measuring physical properties other than the statistical speckle pattern (for example, spatial correlations within the sample), the speckles reduce the signal-to-noise ratio of the examined light scattering so strongly that it may be impossible to extract the desired quantitative information from the experimental data. Therefore, one has to suppress the speckle noise. This can only be achieved by superimposing a number N of statistically independent speckle patterns, which reduces the contrast by a factor of $\sqrt{N}$. Experimentally this can be done in different ways, as already suggested by Goodman[1] and McKechnie.[2]

One possibility is to destroy the spatial or temporal coherence of the light, e.g., by using white light consisting of many frequency components, or to illuminate the surface with many different (mutually incoherent) lasers. These possibilities are experimentally excluded when performing a light scattering experiment.

The second possibility for suppressing the speckle noise is to expand the laser beam or the detector aperture. This reduces the average length scale of the speckle pattern as compared to the size of the aperture, thus performing the spatial averaging in the light recording system. This possibility is disregarded here, because its use is often impractical in a light scattering setup.

What remains is to perform a spatial or temporal averaging over the speckle pattern by making use of the spatially random distribution of the inhomogeneities and the fact that the light recording system is not able to detect the phase information contained in the speckle pattern. The same average is obtained in liquid systems such as polymer solutions, melts, or glasses well above the glass transition, where the speckle noise is suppressed by fast movements of the scatterers.

This averaging process is achieved in solid systems by moving either the sample or the aperture of the light recording system. The latter possibility is excluded experimentally in a light scattering experiment, too. The sample movements can be performed either by rotating the sample around the normal to the scattering plane or by translating it a certain distance perpendicular to the laser beam.

If the scatterers were shifted only inside the beam dimensions, the speckle pattern would remain completely unchanged. On translating the sample, however, new scatterers enter the laser beam while others leave. If there exist no long-range correlations between these scatterers, this consideration yields immediately a necessary translation $\delta = ND$ for obtaining N new speckle patterns produced by a beam of diameter D. This result is confirmed by a more elaborate treatment (see Appendix). On rotating, uncorrelated speckle patterns are generated if the sample is turned through an angle

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$\varphi > 3/Dq$ (see Appendix), with the scattering vector $q = (4\pi/\lambda)\sin(\theta/2)$.

If one inserts some plausible values ($\lambda = 632.8$ nm, $\theta = 90$ deg, $D = 100$ μm), it is found that a shift of approximately 0.2 m is required in order to produce the same number of uncorrelated speckle patterns as obtained from a rotation of 180 deg. This confirms that rotation is a much more efficient way to suppress speckle noise than translation. Nevertheless, both types of movement are described in the literature,[3,4] where mainly the problem of nonergodicity of otherwise isotropic samples has been treated.

However, samples with a preferential direction, for instance uniaxially stretched samples, must be moved in a way that maintains the orientation of this preferential direction with respect to the scattering plane, so that a rotation of the sample is excluded. On the other hand, the geometric dimensions of drawn samples are usually restricted. Therefore, it may well be that one cannot translate the sample sufficiently, say some centimeters. It would be expected, then, that it is not possible to reduce the speckle contrast sufficiently.

In this paper we demonstrate that even translations of only a few times the beam diameter can reduce the speckle contrast considerably. This proves that suitably chosen sample movements are a convenient way to suppress speckle noise in isotropic as well as in uniaxially oriented materials.

2 Experimental Part and Results

A standard light scattering apparatus equipped with a Kr^+ laser ($\lambda = 647.1$ nm) and a goniometer supplied by ALV, Langen, Germany, was used. The lens ($f = 200$ mm) focusing the laser light into the sample produces a spot size of roughly 100 μm at the beam waist. To perform the rotational sample movement, the sample cell was fitted with a rotation stage, consisting of a stepper motor connected to a reduction gear. The vertical movement was achieved by a home-made translation stage driven by a programmable DC-Mike-Drive stepper motor supplied by Physik Instrumente, Waldbronn, Germany (see Fig. 1).

As an example for an isotropic material, the glassy polymer poly(methylmethacrylate) (PMMA) was chosen. The material was supplied by Röhm AG, Darmstadt, Germany, and had a cylindrical shape with 10-mm diameter.

The nonisotropic material was a semicrystalline statistical terpolymer consisting of 52 mol% vinylidene difluoride, 36.5 mol% tetrafluoroethylene, and 11.5 mol% hexafluoropropylene, which was supplied by Hoechst AG, Frankfurt, Germany. This material, abbreviated in what follows by FST, is translucent even in samples of some centimeters depth, due to its low crystallinity of about $\phi_c = 0.1$, and shows an impressive bluish appearance due to Rayleigh light scattering. The melt-crystallized polymer was uniaxially drawn to an extension ratio $\lambda = L/L_0 = 7$, and a cuboid of $5.7 \times 2.4 \times 2.2$ mm^3 was obtained.

The samples were index-matched with silicone oil and ethanol, respectively. The light scattering intensity was normalized with a methylbenzene reference, and only the polarized component, i.e., the laser light polarized perpendicular to the scattering plane with the polarizer on the goniometer having the same orientation, is investigated in this paper.

Figure 2(a) shows the light scattered from the PMMA sample. The error bars mark the standard deviation of five

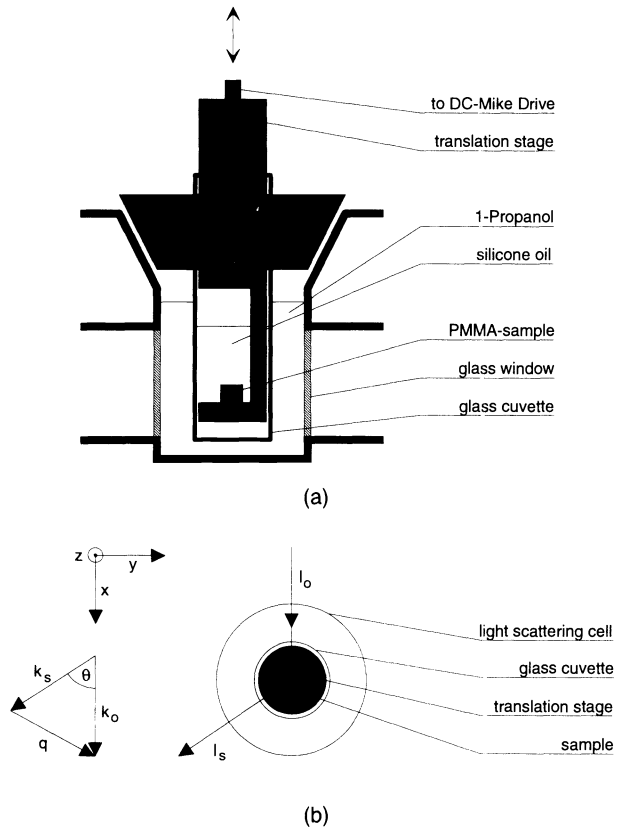


Fig. 1 (a) Experimental setup of the scattering experiment with the translation stage inserted. View in opposite beam direction. (b) Sketch of Fig. 1(a), view from the top. I_0 and I_s are the intensities of incoming beam and stray light, respectively, scattered by an angle θ; $\mathbf{k}_0$ and $\mathbf{k}_s$ denote the accessory wave vectors. The *x-y-z* frame shows the unit directions referred to in the Appendix.

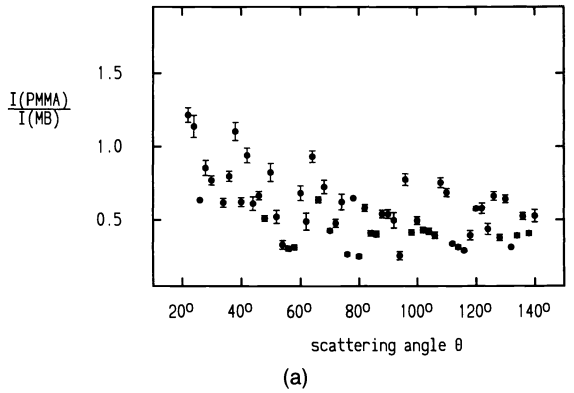
distinct measurements lasting 5 s each. The strong scatter of data points is characteristic for a sample showing a strong speckle effect.

If the same sample is rotated with an angular velocity $\omega = 10\ s^{-1}$ and an integration time $t = 25$ s as above, the scattering curve of Fig. 2(b) is obtained. The characteristic spatial correlation of glassy polymers,[5,6] often termed clusters, is clearly visible, and the speckle noise can no longer be observed. The strong increase of the scattering towards small scattering angles is almost completely obscured by the speckles in Fig. 2(a), in contrast with Fig. 2(b).

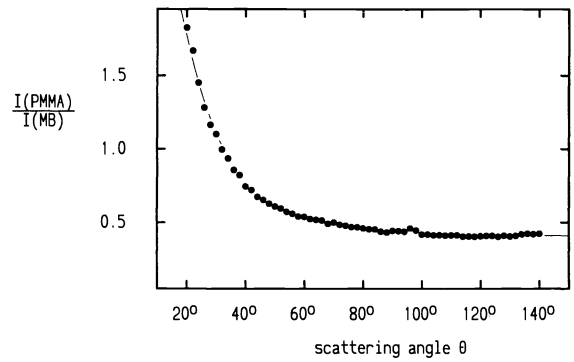
Figure 2(c) shows the result for the sample above with the movement perpendicular to the scattering plane. The DC-Mike drive ran back and forth a distance of 0.3 mm with a constant velocity $v = 0.1\ mm \cdot s^{-1}$. The speckle contrast is not completely reduced to zero, because an insufficient number of uncorrelated speckle patterns were produced over the moving distance. However, the desired correlations are clearly visible. This result can be improved by moving the sample over a larger distance. The result for this distance is shown for comparison with the FST sample.

The spatial correlation observed in glassy polymers is often fitted with a Debye-type correlation function[7]:

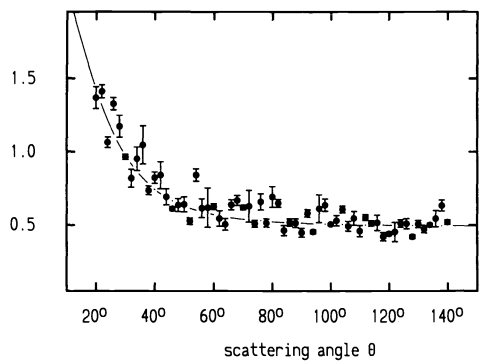
$$\frac{I - I_{BG}}{I_{ref}} = \frac{a}{(1 + q^2\xi^2)^2}\ , \qquad (1)$$



(a)

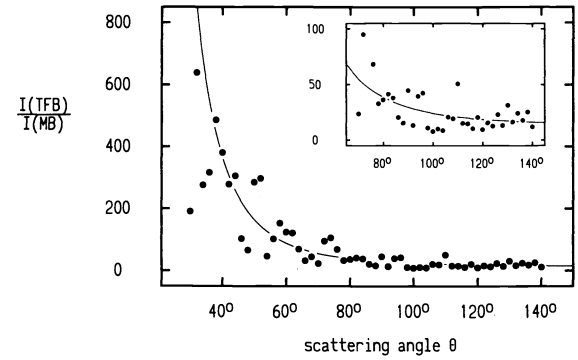


(b)

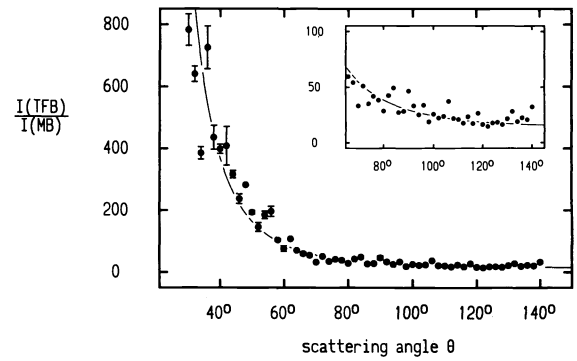


(c)

Fig. 2 Scattered intensity of the PMMA sample versus scattering angle. (a) The sample is at rest and shows a strong speckle pattern. (b) The sample is rotated; the drawn line is a fit of Eq. (1) with $\xi = 135 \pm 1$ nm. (c) The sample is moved along the normal to the scattering plane; the drawn line is a fit of Eq. (1) with $\xi = 135 \pm 7$ nm.



(a)



(b)

Fig. 3 Scattered intensity of the FST sample versus scattering angle. (a) The sample is at rest; the inset shows a detail of the same data at large scattering angles. The straight line is a fit to the data of Fig. 3(b) with Eq. (2). (b) The sample is moved along the normal of the scattering plane; the inset shows a detail of the same data at large scattering angles. The straight line is a fit of Eq. (2).

where I, I_{BG} and I_{ref} denote the measured, the background, and the reference intensity, respectively, $q = (4\pi/\lambda)\sin(\theta/2)$, and a is a constant. A fit of the data of Fig. 2(b) as well as Fig. 2(c) to Eq. (1) results in a correlation length of long-range density fluctuations $\xi = 135$ nm. That the same value is obtained from both data sets demonstrates that in spite of the performed movements the physically relevant parameter can be extracted correctly from the measurements.

Figure 3(a) and 3(b) show the light scattering of the drawn terpolymer sample at rest and with the movement of the translation stage as above. The distance used, 300 μm, was the largest over which the sample could be shifted without dirt particles showing up in the scattering curve. The scattering intensity of the semicrystalline sample is two orders of magnitude higher than for the glassy sample because of the strong optical contrast between crystalline and amorphous domains.

The data of Fig. 3(b) are fitted to the Porod-type behavior[8] of Eq. (2), which here only serves as a guide to the eye:

$$\frac{I - I_{BG}}{I_{ref}} = \frac{a}{q^4} \; ; \tag{2}$$

the symbols have the same meaning as in Eq. (1). Again, the signal-to-noise ratio is strongly improved by the translational movement, as is especially obvious at small scattering angles. This result demonstrates that, even with a translation of only 300 μm (i.e., 3 times the beam diameter in the sample), a considerable reduction of speckle noise can be achieved.

Further investigations of the light scattering behavior of the semicrystalline polymer used in this study will be presented elsewhere.[9]

3 Conclusions

We have demonstrated that suitably chosen movements of solid polymer samples can reduce disturbing speckle noise to an extent that enables a physical interpretation of the information otherwise obscured by the speckle noise, without

compromising the significance of this information. Although a sufficient reduction of speckle noise is expected with movements of some millimeters, it could be demonstrated that it is sufficient to average over only a few speckle patterns to reduce the speckle contrast considerably.

The suitable movement depends on the polymer material and the sample geometry as well. Therefore it remains necessary to examine the kind and extent of motion needed for each sample and material anew.

4 Appendix

The intensity autocorrelation function of the speckles produced by passing the laser beam in the x direction through the sample positions y_1, z_1 and y_2, z_2, respectively, can be shown to be[10]

$$\langle I(y_1,z_1)I(y_2,z_2)\rangle = \langle I(y_1,z_1)\rangle\ \langle I(y_2,z_2)\rangle + |\langle E(y_1,z_1)E^*(y_2,z_2)\rangle|^2$$

with the scattered electric field $E(\mathbf{q}) = \sum_{j=1}^{n} a_j \exp(i\mathbf{q}\cdot\mathbf{r})$. Uncorrelated speckle patterns are characterized then by $\langle E(y_1,z_1)E^*(y_2,z_2)\rangle \approx 0$.

To calculate the field correlation for a sample rotation through an angle φ in the x,y plane (with x now for simplicity chosen to be parallel to $\mathbf{q}$), one gets

$$\begin{aligned}\langle E(x,0)E^*(x,y)\rangle &\sim \langle a^2\ \exp(iq_x x) \times \exp[-i(q_x x\ \cos\varphi + q_y y\ \sin\varphi)]\rangle \\ &\sim \iint \exp\left(-\frac{2x^2}{D^2}\right) \exp\left(-\frac{2y^2}{D^2}\right) \times \exp[iq_x x(1-\cos\varphi)] \times \exp(-iq_y y\ \sin\varphi)\ \mathrm{d}x\ \mathrm{d}y\end{aligned}$$

if the incident laser light is characterized by a Gaussian intensity profile with diameter D, which persists in the scattered radiation. Some straightforward algebra together with an expansion of the angular functions to the lowest order of φ leads to

$$\langle E(x,0)E^*(x,y)\rangle \sim \exp\left(-\frac{(Dq\varphi)^2}{8}\right) ,$$

which immediately yields the condition $\varphi > 3/Dq$ for the small angle through which the sample has to be turned to give decorrelated speckle patterns.

The calculation for the case of translating the sample is somewhat easier. The field correlation function is

$$\begin{aligned}\langle E(\mathbf{r})E^*(\mathbf{r}+\boldsymbol{\delta})\rangle &\sim \langle a(\mathbf{r})\ a(\mathbf{r}+\boldsymbol{\delta})\ \exp(i\mathbf{q}\cdot\mathbf{r})\ \exp[-i\mathbf{q}\cdot(\mathbf{r}+\boldsymbol{\delta})]\rangle \\ &= \langle a(\mathbf{r})\ a(\mathbf{r}+\boldsymbol{\delta})\ \exp(-i\mathbf{q}\cdot\boldsymbol{\delta})\rangle ,\end{aligned}$$

which reduces to

$$\langle E(z)E^*(z+\delta)\rangle \sim \langle a(z)\ a(z+\delta)\rangle$$

if one performs translations $\boldsymbol{\delta}$ parallel to z (which are perpendicular to $\mathbf{q}$). If again a Gaussian intensity profile for the incident laser beam is assumed, the correlation function yields

$$\begin{aligned}\langle E(z)E^*(z+\delta)\rangle &\sim \int \exp\left(-\frac{z^2}{D^2}\right) \exp\left(-\frac{(z+\delta)^2}{D^2}\right) \mathrm{d}z \\ &\sim \exp\left(-\frac{\delta^2}{D^2}\right) ,\end{aligned}$$

which in turn leads to $\delta > D$ for the necessary shift of the sample.

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