Near-field intensity correlations of scattered light

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We show that the two-point correlation function in the near field of scattered light is simply related to the scattered intensity distribution. We present a new, to our knowledge, optical scheme to measure the correlation function in the near field, and we describe a processing technique that permits the subtraction of stray light on a statistical basis. We present experimental data for solutions of latex spheres, and we show that this novel technique is a powerful alternative to static light scattering. © 2001 Optical Society of America

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1. Introduction

When light is scattered by a medium with random fluctuations of the refractive index it produces a speckled field that is due to the random interference of the scattered elementary waves. The statistical properties of the speckle field have been extensively investigated. In particular, it is well known that the space intensity correlation in the far field of the scattered light does not provide any information on the statistical properties of the scatterers because the Van Cittert–Zernike (VCZ) theorem dictates that the equal-time, two-point correlation function depends on only the actual main beam’s intensity distribution. On the other hand, it is well known that the time correlation is simply related to the dynamic properties of the scatterers, and this is the basis of the well-known experimental technique of intensity-fluctuation spectroscopy.

In a previous study, we showed that information on the scatterers may actually be obtained by means of taking the two-point space correlation of the scattered light, provided that this operation is performed in the near field. In particular, we showed that the field correlation is connected with the intensity distribution \( I(q) \) of the scattered light simply through a Fourier transform.

In this paper, we present a new, to our knowledge, optical scheme for measuring the correlation function. With the present configuration a much better instrumental response is obtained. We discuss in detail the requirements that the optical layout must satisfy. We also describe a processing technique that permits the subtraction of stray light on a statistical basis. We present data obtained with solutions of latex spheres with diameters of 5 and 10 \( \mu \)m in water, and we compare them directly with data from low-angle scattering. We show that this novel technique is a powerful alternative to static light scattering and that it offers some distinct advantages, as it allows one to realize an instrumental arrangement that requires minimal optical components and is free of alignment requirements.

2. Theory

To understand the principle of operation of the near-field intensity-correlation technique, we review here the basic results presented in Ref. 4. As a first step, we recall the VCZ theorem. It states that the correlation function of the field scattered by a random sample at a certain distance is simply given, up to multiplicative constants, by the Fourier transform of the intensity distribution of the sample. To put things in a mathematical form, we have

\[
J_A(\Delta x, \Delta y) = \langle E(x_1, y_1)E^*(x_2, y_2) \rangle = \int \int I(\xi, \eta)\exp\left[i\frac{2\pi}{\lambda z}(\xi \Delta x + \eta \Delta y)\right] d\xi d\eta,
\]

(1)

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where \((x_1, y_1)\) and \((x_2, y_2)\) are two points in the observation plane \(x-y\), \(\Delta x = (x_1 - x_2)\), \(\Delta y = (y_1 - y_2)\), \(E\) is the field in the observation plane, \(\lambda\) is the wavelength, and \(I(\xi, \eta)\) is the actual intensity distribution of the sample in the plane \(\xi-\eta\) at a distance \(z\) from the observation plane. In writing Eq. (1), we omitted a proportionality constant.

The theorem holds for \(\delta\)-correlated sources, that is, sources that shine light in any direction (like atoms) and under the assumption that both the linear dimension of the source and the distances \(\Delta x\) and \(\Delta y\) between points in the observation plane are small compared with \(z\). The intensity-correlation function \(R_J(\Delta x, \Delta y) = \langle I(x_1, y_1)I(x_2, y_2)\rangle\) is then derived by the application of the so-called Siegert relation\(^1\)

\[
R_J(\Delta x, \Delta y) = \langle I \rangle^2 + |J_A(\Delta x, \Delta y)|^2,
\]

which holds for fields with circular complex Gaussian statistics.

From Eqs. (1) and (2), we learn that the intensity correlation is related to only the intensity distribution of light incident upon the sample but that it does not contain information about the scatterers. The average size \(\phi\) of a speckle depends on the size of the source \(D\), on the distance \(z\) of the observation plane from the source, and on the wavelength \(\lambda\) as \(\phi = \lambda z/D.\)

Let us now consider a very peculiar source, namely, a beam of diameter \(D\) that illuminates a sample of large particles with diameters of \(d > \lambda\). As is shown in Fig. 1(a), the particles scatter light mainly in the forward direction in a cone with an angular width of \(\Theta = \lambda/D\). In accord with the VCZ theorem, if a sensor is placed far from the sample it sees the speckle field generated by the source of diameter \(D\) and measures an intensity autocorrelation function that does not contain information on the scattering particles. In contrast, if a sensor is placed very close to the sample—we clarify below what “very close” actually means—it receives light from only a portion of the spot with a diameter of \(D^* < D\), as is shown in Fig. 1(b). \(D^*\) is obviously related to \(\Theta\), and we can estimate that \(D^* = \Theta_2 = \lambda z/d\). The brightness of the source, as seen by the sensor, changes with the observation angle in a way that mirrors the intensity distribution of the light scattered by the particles. We can then rewrite the VCZ theorem in a slightly different way by writing the intensity distribution of the effective source in the plane \(\xi-\eta\) as a function of \(r\), a vector whose components are \(q_x = 2\pi \xi/(\lambda z)\) and \(q_y = 2\pi \eta/(\lambda z)\) and that coincides with the scattering wave vector for small scattering angles. With this new definition the VCZ theorem becomes

\[
J_A(r) \propto \int I(q) \exp(i\mathbf{q} \cdot \mathbf{r}) dq,
\]

where \(r = (\Delta x, \Delta y)\).

Therefore we find that the field autocorrelation function in the near field is simply proportional to the Fourier transform of the scattered intensity distribution. By analogy with the far-field case the size \(d_{sp}\) of the speckles in the near field is related to the effective source diameter \(D^*\) through the relation \(d_{sp} \approx \lambda z/D^* \approx d/z\). This result is very interesting as it shows that the speckle field close to the source does contain information about the scatterers and does not depend on the distance \(z\), provided that \(D^* < D\), i.e., that \(z < 4D/\lambda\). Note that the conclusions obtained here with simple and intuitive arguments can be rigorously drawn from the VCZ theorem if one considers the case of a source that is not \(\delta\) correlated.\(^6\)

3. Experiment

In a previous study,\(^4\) we presented data that were obtained with opaque metallic screens that had a random distribution of pinholes of given diameters. The speckle fields were collected by a multielement sensor (an easily obtainable CCD) that interfaced with a PC, and the intensity correlation of the images was calculated through software. We showed that the results of that study hold exactly. In particular, measurements were performed at different distances from the sample, and we also showed that the speckle size does not depend on the distance from the sample and does coincide with the size of the scatterers (pinholes). This conclusion is in sharp contrast to that of the far-field situation in which the speckle size increases linearly with the distance from the sample and there is no relation with the size of the scatterers. The data were also compared with calculated profiles, and the agreement was very good.

The kind of sample used in the earlier study\(^4\) was simple and ideal from many points of view: The scatterers are larger than the pixel size (we used pinholes of 140 and 300 \(\mu\)m in diameter), they are static, and there is no transmitted beam. Actually, in a real scattering experiment the transmitted beam should be removed to measure the space intensity autocorrelation in the near field of the scattered light only. This requirement slightly complicates the optical layout, although it does not change anything from a conceptual point of view. Furthermore, the
scatterers' diameter is usually smaller than—or comparable with—the CCD pixel size (typically 5–10 \( \mu \text{m} \)); therefore an enlarging optical scheme is necessary. Finally, the scattering particles execute rapid diffusive motion, and the acquisition time must be shorter than the shortest time constant associated with the largest scattering wave vector. In Ref. 4, we analyzed all these problems and discussed a preliminary setup in which the transmitted beam was disposed of by our blocking it in the magnifying lens's focal plane. We presented data that were obtained with latex particles of different diameters. The results were quite encouraging, but some nonmarginal problems remained.

In particular, the collecting lens had a limited maximum angle of acceptance, and this restriction introduced a substantial instrumental profile. Also, no attempt was made to deal properly with the problem of stray light. Both of these points are considered in the present study.

In Fig. 2 the optical scheme of the new experimental setup is shown. A parallel beam with a diameter of \( D = 2 \text{ cm} \) is sent into the sample, a 1-mm-thick cell containing latex particles. To reduce the width of the instrumental profile, we use as a collection lens a 20\( \times \) microscope objective, and this lens appreciably increases the maximum acceptance angle.\(^7\) The microscope objective conjugates a plane that is 1 cm past the cell with the plane in which the CCD camera is placed. The transmitted beam is removed by means of a beam stop that is placed in the back focal plane of the entrance block of lenses of the microscope objective.\(^8\)

Measurements were performed with latex spheres of 5 and 10 \( \mu \text{m} \) in diameter. In Fig. 3 the speckle fields obtained with the two samples are shown. The typical sizes of the speckles are very obviously different and roughly a factor of 2 larger for the larger particles, as expected. This result is direct evidence of the speckle dependence on the particles' size and not on the intensity profile of the sample. The number of particles contributing to the field within one speckle can be estimated to be of the order of a few hundred. Therefore the speckle field is Gaussian, and the information about the location of the particles is lost in the images.

To remove the contribution of stray light requires that the data be appropriately processed. This is a nontrivial problem because, in principle, we cannot assume that the stray-light contribution is additive, as is the case with conventional static light scattering. It has been indicated that a substantial fraction of stray light possesses the same statistical properties of the light truly scattered by the sample.\(^9\) In our case this means that both the stray light and the field scattered by the sample are Gaussian. Under

![Fig. 2. Optical layout: A lens (a microscope objective) images a plane immediately after the cell onto a CCD camera. The transmitted beam is removed through a beam stop that is placed in the focal plane of the lens.\(^8\) The lens magnification is the ratio \( q/p \) (not in scale in the figure).]

![Fig. 3. (a) Near-field speckles produced by a sample of latex spheres of 5 \( \mu \text{m} \) in diameter. (b) Near-field speckles produced by a sample of latex spheres of 10 \( \mu \text{m} \) in diameter. Note the difference in the size of the speckles.]

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this condition a good approximation is that the stray-light field correlation sums to the scattered field correlation. Then the scattered field correlation function can easily be recovered if the stray-light field correlation is measured. The near-field intensity contains contributions both from the light scattered by the sample and from stray light. Although light scattered by the sample fluctuates over time, stray light is time independent. Thanks to this difference, the stray-light contribution may be isolated by the averaging of approximately 100 images that are grabbed far apart in time so that the temporal fluctuations are averaged to zero.

The experimental procedure is as follows: A large number of images (typically 100) of the near-field intensity is collected. The intensity-correlation function is calculated for each image, and the average of all these functions, the total intensity correlation \( \langle R_I \rangle_{\text{TOT}}(r) \), is then obtained. The images are averaged, and the intensity correlation \( \langle R_I \rangle_{\text{M}}(r) \) of the resultant image is calculated. Note that \( \langle R_I \rangle_{\text{M}} \) is the correlation of the mean intensity distribution, i.e., the mean intensity of the light scattered by the sample, plus the stray-light speckle field. Finally, for each image, one calculates the mean intensity \( \langle I \rangle \) by summing up the intensity read by each pixel and dividing by the number of pixels; thus the average mean intensity \( \langle \langle I \rangle \rangle \) of the whole set of images is obtained.

Using Eq. (2), we calculate the field autocorrelation function \( J_{\text{TOT}}(r) \) of the total light (the light scattered by the sample and the stray light) as

\[
J_{\text{TOT}}(r) = [(R_I)_{\text{TOT}}(r) - \langle I \rangle^2]^{1/2}
\]

and the field autocorrelation function \( J_{\text{SL}}(r) \) of the stray light as

\[
J_{\text{SL}}(r) = [(R_I)_{\text{M}}(r) - \langle I \rangle^2]^{1/2}.
\]

Finally, the scattered field correlation is obtained as

\[
J_A(r) = J_{\text{TOT}}(r) - J_{\text{SL}}(r).
\]

This way of removing the stray-light contribution represents a great advantage with respect to low-angle light scattering because it is possible to derive from the sequence of near-field images both the stray-light contribution and that resulting from the truly scattered light. At variance with the near-field technique, with static light scattering two different measurements are required: a reference measurement and a measurement of the sample under investigation. Often this way of proceeding cannot be followed or, if followed, leads to improper subtraction.

In Fig. 4, we show the experimental data obtained with latex spheres of 5 and 10 \( \mu \) m in diameter. The scattered intensity distributions \( I(q) \) were obtained by the Fourier transformation, in accord with expression (3), of the field autocorrelation functions calculated from the images, as explained above [see Eq. (6)]. Experimental curves are compared with the scattered intensity distributions from the same samples that were measured by use of a traditional light-scattering instrument.\(^\text{10}\) The agreement is good and shows that the performance of this novel technique is quite comparable with that of low-angle light scattering. In contrast to the results of Ref. 4, here the instrumental response plays a marginal role.

4. Discussion and Conclusions

To compare this novel experimental technique with low-angle light scattering, we find that some further observations about the optical configuration shown in Fig. 2 can be helpful. In a typical light-scattering experiment the intensity distribution is measured in the focal plane of the collection lens. This measure-
ment is usually done either with a sensor designed to remove the transmitted beam\textsuperscript{10} or by the imaging onto a CCD sensor of the focal plane, where a beam stop is placed.\textsuperscript{11} In both configurations there is a direct relation between the scattering angles and the distances from the optical axis. Light scattered at a given angle lies on a circle centered about the focal point of the lens. In the experimental setup shown in Fig. 2 the collection lens images the near-field scattered light onto the CCD sensor. No accurate positioning of the CCD is needed. In fact, at variance with conventional light-scattering techniques is that all the CCD pixels contain information on every scattering angle. Furthermore, moving the CCD along the $z$ direction only changes the plane that the lens conjugates on the CCD, and all the planes are equivalent, provided that their distance from the cell is $z < dD/\lambda$.

In conclusion, we believe that the technique described here is a powerful alternative to low-angle light scattering. It requires a very simple setup, and, thanks to the large number of pixels in a CCD and the good handling capabilities of PCs, it can easily produce smooth high-finesse data. Also, the technique simplifies the problem of stray-light subtraction, possibly the principal problem of any low-angle static light-scattering measurement.

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References and Notes
7. By this we mean that the intensity correlation obtained with a $\delta$-correlated sample is quite narrow.
8. The back focal plane of the entrance block of lenses of the microscope objective lens is inside it; therefore the beam stop was inserted into the objective itself. As a beam stop, we used a piece of a razor blade cut at 45° and lapped accurately. The transmitted beam is reflected and passes through a hole made in the microscope objective.