Light Scattering Probes of Viscoelastic Fluids and Solids
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REVIEW

Light Scattering Probes of Viscoelastic Fluids and Solids

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ABSTRACT

In this article, we discuss recent advances in static and dynamic light scattering applied to soft materials. Special emphasis is given to light scattering methods that allow access to turbid and solid-like systems, such as colloidal suspensions, emulsions, glasses, or gels. Based on a combination of single- and multispeckle detection schemes, it is now possible to cover an extended range of relaxation times from a few nanoseconds to minutes or hours and length scales below 1 nm up to several microns. The corresponding elastic properties of viscoelastic fluids or solid materials range roughly from below 1 Pa to several 100 kPa. Different applications are discussed such as light scattering from suspensions of highly charged colloidal particles, colloid and protein gels, as well as dense surfactant solutions.

Key Words: Dynamic light scattering; Photon correlation spectroscopy; Diffusing wave spectroscopy; Microrheology; Colloids; Biopolymers; Micelles.

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INTRODUCTION

To characterize the structural and dynamic properties of soft materials, information on the relevant mesoscopic length scales is required. Such information is often obtained from traditional photon correlation spectroscopy (PCS) techniques, such as static and dynamic light scattering (SLS/DLS) in the single-scattering regime. In dense systems, such as colloidal suspensions and gels, however, these powerful techniques frequently fail due to strong multiple scattering of light. Furthermore, many (dense) soft materials are viscoelastic solids, meaning that they are nonergodic. Ergodicity, however, is an important condition for the applicability of PCS.

We will review some of the recent advances in light scattering from turbid fluid- and solid-like media. After a brief introduction to dynamic light scattering, we will show how multiple scattering suppression schemes are used to study moderately turbid systems. These techniques provide the same kind of information as conventional SLS/DLS, even in the presence of significant multiple scattering. For even more turbid samples, diffusing wave spectroscopy (DWS) can be used in order to study the internal dynamics by measuring the intensity fluctuations of the diffusively transmitted light. Because in the case of DWS the light is scattered from a large number of scatterers, each individual one must move only a small fraction of a wavelength for the cumulative change in optical path length to be a full wavelength. Therefore, DWS can probe motion on very short length scales from less than 1 nm to about 50 nm. In the second part of this article, we address in detail how to study the internal dynamics of solid-like nonergodic materials in the single and multiple scattering regimes. Finally, we will illustrate our considerations with a number of recent experiments on soft complex materials, such as correlated colloidal suspensions and gels. Most of our examples were taken from studies on colloidal particle assemblies, where multiple scattering and nonergodicity frequently play an important role. However, our conclusions can easily be applied to any kind of soft material, provided the relevant length and time scales are accessible to light scattering.

PHOTON CORRELATION SPECTROSCOPY

Coherent laser light scattered from a rough or disordered material shows the typical random speckle pattern. If the material under investigation is soft, the individual speckles fluctuate due to the microscopic motion in the medium. Photon correlation spectroscopy analyzes these temporal fluctuations with a time resolution down to a few nanoseconds. Based on the experimentally obtained intensity autocorrelation function (ICF), local dynamic properties on length scales from one to a few hundred nanometers can be studied. Many different methods and geometries are employed for this kind of experiments. A selection of current experimental techniques will be discussed in this section.

Dynamic Light Scattering (DLS) from Weakly Scattering Materials

In a dynamic (single) light scattering (DLS) experiment, laser light (wave vector $\mathbf{k}$) is incident on a scattering cell of dimension $L$ containing a weakly scattering medium.
This means that in the case of conventional DLS, the scattering mean free path \( l > L \) and, therefore, the transmission in line of sight has to be high, \( \exp(-L/l) > 95\% \). In the most simple case, we can consider a system of \( N \) independent point scatterers illuminated with laser light. The scattered light is then detected at a certain angle \( \theta \); hence, incoming and scattered wave vector (with \( |k| = |k'| \), elastic scattering) define the scattering vector \( q = k - k' \) (Figure 1). Each scatterer at a position \( r_i \) feels the same incident field amplitude:

\[
E(r_i) = E_0 e^{ik(r_0 + r_i) - \omega t} = (E_0 e^{-i\omega t}) e^{ikr_i}
\]  

(1)

This field amplitude induces a dipole of the same frequency emitting a spherical wave. Hence, the field amplitude at the detector position is given by the following:

\[
E_s \propto E_0 e^{ikr_i} \sum_{N} e^{i q r_i(t)}
\]  

(2)

In the far field (\(|r| \gg |r_i|\)):

\[
|r - r_i| \approx r - r_i \frac{k'}{|k|} + \cdots
\]  

(3)

and we obtain the following expression for the field amplitude at the detector position:

\[
E_s(q, t) \propto \frac{e^{i kr}}{r} \sum_{i=1}^{N} e^{i(k-k')r_i(t)} = \frac{e^{i kr}}{r} \sum_{i=1}^{N} e^{i qr_i(t)}
\]  

(4)

To experimentally characterize the internal dynamics, the intensity autocorrelation function (ICF) is introduced:

\[
G_2(q, t) = \langle I(q, 0) I(q, t) \rangle_T
\]  

(5)

\[\text{This approximation for weak scattering is equivalent to the first Born approximation. Within the frame of the Raleigh Gans Debye (RGD) formalism, it can easily be expanded to scatterers of any size } a \text{ and shape, as long as the scattering contrast is weak } (ka|n_1/n_2 - 1| \ll 1, \text{ where } n_1/n_2 \text{ denotes the index contrast}).[16,17]\]
For ergodic (fluid-like) systems, the normalized ICF can be directly related to the normalized field autocorrelation function, $g_1(t)$, via the Siegert relation:

$$g_2(q, t) = \frac{\langle I(q, 0)I(q, t) \rangle_T}{\langle I(0) \rangle_T^2} = 1 + \beta |g_1(q, t)|^2$$  \hspace{1cm} (6)

$$g_1(q, t) = \frac{\langle E(q, 0)E(q, t) \rangle_T}{\langle |E(q, 0)|^2 \rangle_T}$$  \hspace{1cm} (7)

The intercept $\beta$ depends primarily on the detection optics and takes an ideal value of 1 that can almost be reached when using single-mode fibers for the detection.\[18\] For randomly moving scatterers, the individual particle mean squared displacement $D_r(t)$ is a random Gaussian variable and Eq. 7 is readily evaluated, assuming that the cross terms $i \neq j$ vanish for uncorrelated scatterers:

$$g_1(q, t) = \frac{\langle E(q, 0)E^*(q, t) \rangle}{\langle |E(q, 0)|^2 \rangle} \propto \sum_{i,j} \langle e^{i\mathbf{q} \cdot \mathbf{r}_i(0) - i\mathbf{q} \cdot \mathbf{r}_j(t)} \rangle \propto \langle e^{-i\mathbf{q} \cdot \Delta \mathbf{r}(t)} \rangle$$

$$= e^{-q^2 \langle \Delta \mathbf{r}(t)^2 \rangle / 6}$$  \hspace{1cm} (8)

Combined with the Stoke–Einstein relation ($D_0 = kT/6\pi \eta a$) for freely diffusing spheres [$\Delta \mathbf{r}(t)^2 = 6D_0t$],\[19\] this expression serves as the basis for the size analysis (particle radius $a$) in the submicron range, one of the most prominent applications of PCS.\[8\]

$$g_1(q, t) = e^{-D_0 q^2 t}$$  \hspace{1cm} (9)

For interacting and therefore correlated scatterers (spherical particles of size $a$) the scattered intensity factorizes: $I(q) = P(q) \times S(q)$. Here, $P(q) = k_0^2 \left( \frac{d\sigma_s}{d\omega} \right)$ is the Mie scattering function of the individual particle.\[16,17\] The intermediate scattering function

$$S(q, t) = S(q) \times g_1(q, t)$$  \hspace{1cm} (10)

then comprises information about structural correlations $S(q)$ and temporal fluctuations via $g_1(q, t)$.

3DDLS and TCDLS—Multiple Scattering Suppression Techniques

While we have seen that SLS and DLS provide interesting insight into the structural and dynamic properties of soft materials, their application to many systems of scientific and industrial relevance has often been considered as too complicated, due to the very strong multiple scattering frequently encountered. The interpretation of a SLS/DLS experiment becomes exceedingly difficult for systems with nonnegligible contributions from multiple scattering. Particularly for larger particles with high scattering contrast, this limits the technique to very low particle concentrations, and a large variety of systems are, therefore, excluded from investigations with dynamic light scattering. However, Schützel demonstrated that it is possible to suppress contributions
from multiple scattering from the measured photon correlation data.\cite{20} During the last few years, a number of different theoretical and experimental approaches to this problem appeared.\cite{21} The general idea is to isolate singly scattered light and suppress undesired contributions from multiple scattering in a dynamic light scattering experiment (for details, see Ref. \cite{20}). This can be achieved by performing two scattering experiments simultaneously on the same scattering volume (with two laser beams, initial wave vectors $k_{i1}$ and $k_{i2}$, and two detectors positioned at final wave vectors $k_{f1}$ and $k_{f2}$) and cross-correlating the signals seen by the two detectors. If both experiments then have the same scattering vector (i.e., in magnitude as well as in direction), but use different scattering geometries, and in the absence of multiple scattering, each detector sees the same spatial Fourier component of the sample. The corresponding relations between the autocorrelation function $g_1(q,t)$ and the measured intensity autocorrelation $G_{11}(t)$ and cross-correlation $G_{12}(t)$ function, where (1) indicates singly scattered photons, can then be written as

$$G_{11}(t) = I_{11}^{(1)}(1 + \beta_{11}|g_1(q,t)|^2)$$ \hspace{1cm} (11)

and

$$G_{12}(t) = I_{12}^{(1)}(1 + \beta_{12}|g_1(q,t)|^2)$$ \hspace{1cm} (12)

In the autocorrelation experiment, the intercept $\beta_{11}$ depends primarily on the detection optics and takes an ideal value of 1 that can be almost achieved when using single-mode fibers for the detection, whereas for the case of the cross-correlation experiment, $\beta_{12}$ is, in addition, reduced due to phase mismatch $\delta q = |\delta q|$ (describes the match of the scattering vectors $q_1$ and $q_2$) and misalignment $\delta x = |\delta x|$ (the spatial match of the scattering volumes). For single scattering, both auto- and cross-correlation, therefore, yield the same information. However, in the case of multiple scattering, the situation changes. In the autocorrelation experiment, the multiply scattered photons contribute to $G_{11}(t)$ as well and make an interpretation of the data and a deduction of $g_1(q,t)$ very difficult, if not impossible. However, in the cross-correlation experiment outlined above, only singly scattered light will produce correlated intensity fluctuations on both detectors. In contrast, multiply scattered light will result in uncorrelated fluctuations that contribute to the background only due to the fact that it has been scattered in a succession of different $q$-vectors, and the contributions from multiple scattering to the signal are suppressed by a factor of order $R\delta k_j/C_0^2$, where $R$ is the beam waist, and $\delta k_j/C_0$ denotes the magnitude of the smallest of the two wave vector combinations $k_{i2} - k_{i1}$ and $k_{i2} + k_{i1}$.\cite{20} This leads to the following expression for $G_{12}(t)$:

$$G_{12}(t) \approx I_1 I_2 + \beta_{12} I_{11}^{(1)} I_{12}^{(1)} |g_1(q,t)|^2$$ \hspace{1cm} (13)

where $I_j$ is the average intensity summed over all contributions (singly and multiply scattered photons) measured at detector $j$, and $I_j^{(1)}$ contains the singly scattered light only. Such a cross-correlation experiment thus provides us with $g_1(q,t)$ even for turbid suspensions, and multiple scattering will be visible in the decreasing intercept only.

Several different proposals for an experimental realization of this task were presented in the past. Schä tzel\cite{20} and his collaborators successfully implemented the
two-color method (TCDLS).\textsuperscript{[22]} While this method has been demonstrated to work very well by several groups (see Ref. [21] and references therein), it is technically extremely demanding and requires sophisticated alignment procedures. A particularly interesting scheme is the so-called three-dimensional cross-correlation experiment (3DDLS), in which the two incident and the two detected light paths are placed at an angle $\delta/2$ above and below the plane of symmetry of the scattering experiment.\textsuperscript{[20,21,23–25]} The initial $(k_{11}, k_{22})$ and final $(k_{13}, k_{23})$ wave-vector pairs are rotated by some angle about the common scattering vector $q = q_1 = q_2$ for the two scattering processes 1-1 and 2-2, whereas the two other scattering processes 1-2 and 2-1 detected in this experiment have different scattering vectors. These additional scattering processes will, therefore, contribute to the background only, which means that the maximum intercept for 3DDLS is only one quarter of the value obtained for autocorrelation or other cross-correlation schemes, such as the two-color method, i.e., $\beta_{12, \text{ideal}} = 0.25$.\textsuperscript{[20]}

A schematic layout of a 3DDLS experiment is shown in Figure 2. A laser beam is split into two parallel beams that are then focused onto the scattering cell by a lens. The lens has to be chosen in order to allow for a sufficiently large angle $\delta$ required for an efficient suppression of contributions from multiple scattering.\textsuperscript{[25]} An identical lens is used for the detection side, and the scattered light with wave vectors $k_{11}$ and $k_{22}$ is collected using single-mode fibers. For turbid colloidal suspensions, due to the strong attenuation of the singly scattered intensity that essentially decays like $I(1) \sim \exp(-s/l)$, the size of the scattering cell becomes important ($s$ is the optical path across the cell for singly scattered light, and $l$ is the scattering mean free path, which may easily be less than 1 mm for dense colloidal suspensions). In order to avoid a strong reduction of the singly scattered light and the subsequent reduction in signal (see Eq. 8), the light path should be of the order of $l$. In principle, this could be achieved best by using flat cells. However, because these cells will then be used in general with beams at nonnormal incidence angles, this will lead to considerable beam deflection and

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**Figure 2.** Schematic view of a 3DDLS setup. Side-view) Two parallel beams are focused onto the scattering cell (vat) by a lens. An identical lens is used for the detection side, and the scattered light is collected using single-mode fibers (SMF). Top-view) A schematic description of the $\theta-2\theta$ geometry for two different scattering angles, angles a) $\theta=90^\circ$ and b) $\theta=30^\circ$, that can be used to work with rectangular scattering cells to minimize the optical path length.
displacement. This problem can be overcome by implementing a different approach, as outlined in Figure 2. One can use square cells with 10 mm path length that are of superior optical quality for experiments at low scattering angles, and position them such that the scattering volume is located in a corner of the cell to have short optical path lengths. For experiments at scattering angles different from 90°, we can then turn the cell in a so-called δ - 2θ geometry, where the sample is rotated by half the rotation angle (90° - θ) in order to recover a symmetrical situation in which the displacement of the incident and scattered beam almost cancel. [23]

A 3DDLS experiment cannot only be used to determine the dynamic structure factor for turbid solutions, but one can also perform a static light scattering experiment, where the 3DDLS scheme allows to correct for multiple scattered light. [21,25] The basis for this is the fact that the intercept of the cross-correlation function depends on the ratio between the singly scattered light I\(_{(1)}(q)\) and the total scattered intensity I(q) (Eq. 8). The mean intensity measured in a static light scattering experiment contains contributions from singly scattered and from different orders of multiply scattered light. To extract the single scattering contribution I\(_{(1)}(q)\), I(q) has to be corrected with the reduced intercept \(\beta_{12}/\beta_{12}^{(1)}\) of the cross-correlation function, where \(\beta_{12}^{(1)}\) denotes the intercept measured in the limit of single scattering only, which can be measured independently in a dynamic light scattering experiment with a highly diluted sample. The single scattering contribution to the total intensity observed at detectors 1 and 2 can then be obtained from

\[
I_{(1)}(q) = \sqrt{I_{1}^{(1)}(q)I_{2}^{(1)}(q)} = \sqrt{I_{1}(q)I_{2}(q)}(\beta_{12}/\beta_{12}^{(1)})
\]

When doing static light scattering measurements with rectangular cells on concentrated suspensions, where the mean free path length \(l\) is of the order of the path length \(s\) in the cell, a transmission correction of the scattering intensity with the scattering angle \(\theta\) is necessary, because \(s\) varies with \(\theta\) (see Figure 2).

Several research groups [21] demonstrated the feasibility of such an experiment and clearly showed that static and dynamic light scattering need not be restricted to dilute suspensions of small particles, but can be used to successfully characterize turbid suspensions with transmission values as low as \(T \approx 0.01\) (which for square cells and \(\delta - 2\theta\) geometry allows for a scattering mean free path of approximately 50 μm or more). This opens a completely new field of suspension characterization using static and dynamic light scattering experiments combined with novel cross-correlation schemes in numerous areas of soft condensed matter research and technology.

**Diffusing Wave Spectroscopy—DWS**

In conventional DLS (Figure 3), the sample has to be almost transparent (and, hence, often highly diluted). As we have seen previously, 3DDL allows some of the shortcomings of DLS to be overcome. However, the signal strength decreases roughly exponentially along with increasing turbidity (or decreasing scattering length \(l\)), and hence, multiple scattering suppression is best applied to moderately turbid samples.

Diffusing wave spectroscopy (DWS) extends the previously described single light scattering techniques (DLS, TCDLS, 3DDL) to strong multiple scattering, by treating...
the transport of light as a random walk (Figure 4). Over the last 15 years, the DWS technique has been applied to study and characterize a variety of optically dense complex systems such as colloidal dispersions and gels, ceramic slurries and green bodies, biopolymers and gels (yogurt and cheese), granular media, and foams.

To derive the autocorrelation function for diffuse transport, we first consider an individual multiple scattering path with $n$ scattering events. The normalized field autocorrelation function of a path of length $s = nl$ is given by

$$g_n(t) = \left[ \int_0^{2k_0} qP(q)S(q,t) dq \right]^n / \left[ \int_0^{2k_0} qP(q)S(q) dq \right]^n$$

with the normalized field autocorrelation function given by

$$g_1(t) \propto \sum_{n=1}^{\infty} P(n)g_n^\alpha(t)$$

In many cases, the intermediate scattering function at sufficiently short times is dominated by a diffusion-type process and can be written the following way:

$$S(q,t) = S(q)e^{-2\alpha t/\beta(q)q^2}$$

In the following, some typical cases are listed:

- For an uncorrelated suspension $S(q) \equiv 1$ of identical particles in a simple liquid:

$$\alpha(t) = D_0t; \beta(q) = 1$$

- For an uncorrelated (dilute) suspension $S(q) \equiv 1$ of identical particles suspended in a viscoelastic fluid or solid:

$$\alpha(t) = \langle \Delta r^2(t) \rangle / 6; \beta(q) = 1$$
For a correlated suspension \( S(q) \neq 1 \) of identical particles:

\[
\begin{align*}
\alpha(t) &= D_0 t; \\
b(q) &= H(q)/S(q)
\end{align*}
\]

For a fractal gel of identical particles in a simple fluid:

\[
\begin{align*}
\alpha(t) &= \delta^2(1 - e^{-(t/c)})/6; \\
b(q) &= 1
\end{align*}
\]

The time-dependent parameter \( \alpha(t) \) characterizes the self-motion of the individual particles as expressed by their mean squared displacement. For small particles and strongly correlated suspensions, collective motion is detected by light scattering. \( b(q) \) takes account for this influence in the examples given above.

Due to the typically large number of scattering events \( n \) along a multiple scattering path, for small values of \( 1 - S(q, t)/S(q) \), a full decay of the DWS correlation function \( g_1^n(t) \) will be observed and, therefore,

\[
S(q, t) \approx S(q)[1 - q^2 \alpha(t) b(q)]
\]  

(18)

hence,

\[
g_1^n(t) = \left[ 1 - \frac{\int_0^{2\kappa} q^3 P(q)S(q)[\alpha(t) b(q)]dq}{\int_0^{2\kappa} q P(q)S(q) dq} \right]^{n}
\]  

(19)

**Figure 4.** DWS setup: An intense laser beam (Verdi from Coherent) is scattered from a turbid sample contained in a temperature controlled water bath. Two-cell DWS (TCDWS): Light transmitted diffusively from the sample cell is imaged via a lens onto a second cell (containing a highly viscous colloidal suspension of moderate optical density). Subsequently, the light is detected with a single-mode fiber (SMF) and analyzed digitally (correlator and PC). Multispeckle DWS (MSDWS): In backscattering, the fluctuations of the scattered light are analyzed with a CCD camera (or a SMF). MSDWS can also be applied in transmission geometry, in parallel to TCDWS, by placing a beam splitter between the sample and the lens.
which we can write again as an exponential (with $e^{-x} \approx 1 - x$, $x \ll 1$):

$$g_1(t) = \exp \left\{ -T_0 \int_0^{\beta_0} q^3 P(q) S(q) [\beta(t) \beta(q)] dq \right\}$$

(20)

We find

$$g_1(t) = \exp \left\{ -2 \beta_0 \frac{s}{l^*} \beta(t) \frac{[\beta \cdot S]}{[S]} \right\}$$

(21)

using $n = s/l^* \times l^*/l$ and the well-known relation between $l$ and $l^*$.\[26,27]

$$\frac{l^*}{T} = 2 \beta_0 \frac{s}{l^*} \beta(t) \frac{[\beta \cdot S]}{[S]}$$

(22)

The transport mean free path $l^*$ is the effective step length of the random walk. (Note that $l^*$ directly determines the transmission coefficient for diffuse light: $T \equiv \frac{I_{transmitted}}{I_{incident}} \approx l^*/L$.) The brackets $[ ]$ in Eq. 21 denote the angular average of the dynamic and static quantities accessible by DWS.\[36,37]

$$[X] = \int_0^{2 \beta_0} q^3 P(q) X(q) dq / \int_0^{2 \beta_0} q^3 P(q) dq$$

(23)

Replacing the sum by an integral, we obtain the following expressions for the field autocorrelation function.\[b] We note again that for an uncorrelated suspension $S(q) \equiv 1$ of scatterers $[\beta \cdot S]/[S] = 1$.

$$g_1(t) = \int_0^\infty P(s) \exp \left\{ -2 \beta_0 \frac{s}{l^*} \beta(t) \frac{[\beta \cdot S]}{[S]} \right\} ds$$

(24)

For transmission through a slab (plane wave limit):

$$g_1(t) \approx \frac{(L/l^* + 4/3) \sqrt{6 \beta_0^2 \gamma(t) [\beta \cdot S] / [S]}}{\sinh [(L/l^* + 4/3) \sqrt{6 \beta_0^2 \gamma(t) [\beta \cdot S] / [S] }]}$$

(25)

\[b\] Absorption can be taken into account by substituting $2 \beta_0 \gamma(t) [\beta \cdot S]/[S] (s/l^*) \rightarrow 2 \beta_0 \gamma(t) [\beta \cdot S]/[S] (s/l^*) + (s/3l_a)$, where $l_a$ is the microscopic absorption length along the multiple scattering paths. For nonabsorbing scatterers, $l_a$ is the absorption length of the pure solvent.
For backscattering from a semi-infinite medium ($L/l^b \rightarrow \infty$, plane wave limit):

$$g_1(t) = \exp \left( -\sqrt{6k_0^2 z(t) \frac{|\beta \cdot S|}{|S|}} \right)$$

(26)

DWS is able to provide information about the local dynamics of particle dispersions, similar to DLS and 3DDLs, however, without any restrictions on particle concentration and turbidity. Due to the typically large number of scattering events, DWS is sensitive to small local displacements in the nanometer range. Therefore, in principle, DWS is well suited for studying solid materials such as colloid or polymer gels, provided the scattering signal is properly analyzed. In the following, we will describe how to do such an analysis in order to access the dynamics of viscoelastic solid materials with PCS.

**NONERGODICITY IN LIGHT SCATTERING**

In solid-like media, the scatterers are localized near fixed average positions, probing only a small fraction of their possible spatial configurations by thermal motion. As a consequence, the measured time-averaged quantities (such as the scattered intensity or its autocorrelation function) differ from the ensemble-averaged ones.[38–42,48] Experimentally, one finds that a series of (time-averaged) measurements on a given sample yields a set of different results, each being of limited use for the characterization of the medium (see inset Figure 11). A comprehensive analysis of nonergodicity in dynamic (single) light scattering was given by Pusey and van Megen in 1989.[38] In the following, we will briefly summarize their main results. The authors consider a viscoelastic solid material, as shown in Figure 5, where the motion of the scatterers is restricted in space (as it is in the case, e.g., for a polymeric or a colloidal gel). Light scattered from such a nonergodic system comprises a fluctuating part $I_F$ and

![Figure 5](image)

*Figure 5.* Left: In a viscoelastic solid (as shown for a colloidal network), the individual particle motion is spatially constrained, leading to a plateau value in $\langle \Delta r^2(t) \rangle$. Right: Due to the constrained motion, the (properly ensemble averaged) correlation function $g_1(t)$ does not decay to zero.
a time-independent part \( I_C \). The time average now depends on the exact configuration of the system: 
\[
\langle I \rangle_T = \langle I_F \rangle_T + I_C.
\]
For a given configuration, the (time-averaged) ICF is given as follows:
\[
g_2(t)_T = \frac{\langle 0(t)_T \rangle}{\langle I \rangle_T^2} = 1 + \frac{\langle I \rangle E^2}{\langle I \rangle T} [g_1(t)^2 - g_1(\infty)^2] + 2 \frac{\langle I \rangle E}{\langle I \rangle T} \left( 1 - \frac{\langle I \rangle E}{\langle I \rangle T} \right) \\
\times [g_1(t) - g_1(\infty)]
\]
(27)

The measured ICF still decays to one for large \( t \), however, never attains a value of 2 at \( t = 0 \), except for in the limiting case of an ergodic medium, where \( g_1(\infty)^2 = 0 \). Illustrated in Figure 5 is how the properly ensemble averaged field correlation decays for a viscoelastic solid. If \( \langle I \rangle_T = \langle I \rangle E \), the second term in Eq. 27 is zero, and consequently, the measured correlation function is simply \( 1 + g_1(t)^2 \) shifted by its plateau value \( g_1(\infty)^2 \). However, in general, the second term in Eq. 27 leads to a complex relation between the measured ICF and \( g_1(t)^2 \) that cannot be solved without prior knowledge of \( \langle I \rangle E / \langle I \rangle_T \).

The Pusey van Megen Method

Pusey and van Megen\cite{38} proposed to use their Eq. 27 to obtain \( g_1(t) \) from a combination of static and dynamic experiments. The idea is to measure the time-averaged ICF (as obtained from a digital correlator) for a single sample orientation, and to subsequently rotate/translate the sample rapidly in order to obtain the ensemble averaged intensity \( I_E \) for a given scattering angle.\cite{38–40,42} This approach can be applied to DLS and DWS.\cite{43} However, it implies that the measured intensity is constant over the measurement time (which is not always easy to achieve with the required precision, in particular for stretched decays or additional slow relaxations).

For multiple scattering suppression techniques, such as TCDLS or 3DDLs, the situation is more complex, because the cross correlated intensity correlation function is analyzed. Here, the nonfluctuating part of the scattered intensity \( I(k_1) \neq I(k_2) \) is not the same for the two cross-correlated signals at directions \( k_1, k_2 \), hence, adding significant complexity to the application of the Pusey van Megen method for these systems. Even if a similar analysis should be possible, one would still suffer a significantly decreased signal-to-noise ratio due to the nonequal count rates at both detectors.c

Rotation and Translation

The most direct method of performing ensemble averaging of scattered light for nonergodic samples is based on the idea of collecting light scattered by different

\[c\text{We note that it seems feasible, though tedious, to manually adjust the cell position in order to have the same signal count rate on both detectors. Under that premise, we assume that the Pusey–van Megen method can also be applied to 3DDLs, though the details of such an approach have not yet been worked out.}\]
parts of the sample, thus performing the "real" ensemble averaging. Experimentally, the sample is slowly moved or rotated, while the ICF is collected. Obviously, this leads to an additional decay of $g_2(t)$, which becomes increasingly rapid with increase of the translation velocity or the rotation frequency. The rotation/translation method can be extended to 3DDLS/TCDLS as well as to DWS without any particular difficulties. It has, however, an important disadvantage of experimental complexity (precise translation or rotation of the sample is required). Also, it has been claimed (based on experimental observations) that this method is less efficient than, e.g., the method of Pusey and van Megen described in the previous section.

**Two-Cell Technique**

To overcome the problem of nonergodicity in DWS, a system of two independent glass cells [thickness $L_1$, $L_2$] can be used. The first cell contains the sample to be investigated, which can be a stable ergodic or an arrested nonergodic sample. The second cell, which serves to properly average the signal of the first cell only, contains an ergodic system with very slow internal dynamics and moderate turbidity (Figure 6). If colloidal particles with $ka > 1$ are used, typical values $L_2 \cong (1 - 2)t^s$ are sufficient to suppress transmission of unscattered light (because $t^s \gg t$).
Sandwich Geometry

The original setup, as described in Refs. [29,48], is based on a sandwich of two glass cells of equal dimensions. Light is incident on the first cell $L_1$. The transmitted speckles at the back of $L_1$ are then diffusively broadened by the presence of the second cell to a size $L_2$. Because the second layer is ergodic, a two-dimensional average over all speckle spots within the same area $L_2^2$ is obtained. The correlation function $g_2(t) - 1$ of the two-cell setup can be expressed through the joint distribution function $P_2(s_1, s_2)$ of path segments $s_1, s_2$ in the cells:

$$g_2(t) - 1 = \left| \int_0^\infty ds_1 \int_0^\infty ds_2 P_2(s_1, s_2) \exp \left\{ -\frac{1}{3} k_1^2 \langle \Delta r_1^2(t) \rangle \frac{s_1}{l_1^*} \right\} \times \exp \left\{ -\frac{1}{3} k_2^2 \langle \Delta r_2^2(t) \rangle \frac{s_2}{l_2^*} \right\} \right|^2$$

(28)

In the case that both layers decouple (i.e., no loops of the scattering paths between the two layers), $P_2(s_1, s_2)$ reduces to a product of two terms $P_1(s_1)P_1(s_2)$ and, therefore, $g_2(t) - 1 = [g_2(t, L_1) - 1][g_2(t, L_2) - 1]$. (A general theoretical treatment of the two-cell technique is given in Refs. [48,51].) One can now directly determine the contribution of the first cell by dividing the signal of the two-cell setup $g_2(t) - 1$ with the separately measured autocorrelation function $g_2(t, L_2) - 1$ of the ergodic system in the second cell:

$$g_2(t, L_1) - 1 = [g_2(t) - 1]/[g_2(t, L_2) - 1]$$

(29)

Separated Cells

Recently, a slightly modified version of the two-cell method was installed by several groups (J. L. Harden, unpublished). In this version, the two cells are separated by a distance of several centimeters, thereby allowing additional optical elements (such as a beam splitter) to be introduced between both cells. The speckle pattern at the surface of cell $L_1$ is imaged via a lens onto cell $L_2$, as shown schematically in Figure 4 (and explained in detail in Ref. [50]). In addition to being more flexible, this setup largely simplifies the decoupling of light propagation in both cells.

Mechanical Translation

Finally, it is worthwhile to mention that efficient averaging can also be achieved by mechanical translation of a solid second cell (such as a glass frit). Obviously, such a setup lacks some of the simplicity of the original two-cell setup. However, if stability with respect to drifts and temperature is required over an extended duration of hours or days, for example for the study of colloidal aging, this can be the method of choice. Also, the decay time of $g_2(t, L_2)$ can be chosen at will and be adapted to the time evolution of the system under study.
Multiple Runs

Another practical way to obtain ensemble-averaged correlation functions is to record a sufficient number of statistically independent time-averaged ICFs. From Eq. 27 with $\langle f \rangle_E / \langle f \rangle_T = 1$:

$$\langle g_2(t)_T \rangle - 1 = \lim_{N \to \infty} \frac{1}{N} \sum_{n=1}^{N} \langle g_2(t) \rangle_T = \frac{g_1(t)^2 - g_1(\infty)^2}{g_1(t)-1}$$

This means that the average of (statistically independent) experimental ICSs yields the correct correlation function $g_2(t) - 1$ shifted by its plateau value $g_2(\infty) - 1$. This result is physically intuitive, because the loss of ergodicity also implies a lower intercept value. It is, therefore, always a strong indication of nonergodicity when the measured intercept value decreases before the previously mentioned reproducibility issues become evident. To ensure statistically independent measurements, it is again possible to measure different system configurations, e.g., by moving the sample.

Sometimes the arrested system can help fulfill this requirement. In many cases, soft solid materials only appear nonergodic but still show a very slow relaxation process, such as aging. While in DLS aging leads to a slow decay on a time scale $\tau_A$ of hours or days, it can be substantially faster in DWS, because the latter technique is more sensitive to subtle changes in the system configuration. Hence, if $\tau_A$ is substantially larger than the time window of interest (e.g., a time window from 10 ns to 1 ms like in Figure 11) but of the order (or slightly larger) compared to the measurement time (typically a few minutes), it is possible to get a reasonable ensemble average $\langle g_2(t)_T \rangle_E$ from a repeated measurement of $g_2(t)_T$. In order to make this method work in practice, one has to adjust the measurement time to $\tau_A$. If the measurement time chosen is too long, the intensity $I_C(q)$ is no longer constant over the time of the measurement, and the digital correlator picks up the slow fluctuations leading to sudden jumps in the experimentally observed correlation function. While this method is certainly less accurate compared to the previous ones, it nevertheless can rapidly provide information about a new material under study.

Multispeckle Analysis

Statistically independent correlation functions are most easily obtained using a CCD camera (see Figure 4). Instead of analyzing the fluctuations of the intensity at a single spatial position (one speckle spot), one analyzes a large area of the intensity pattern of the scattered light (hence, multispeckle) using a CCD camera. Such an approach was first used in dynamic light scattering to replace a standard detector in a goniometer SLS/DLS setup. Later, it was demonstrated that, particularly for low-angle SLS/DLS, an area CCD detector has tremendous advantages for the study of slow relaxation processes. Multispeckle DWS (MSDWS) was reported in 1993, it is relatively simple to set up due to the weak angular dependence of the diffuse light, and furthermore, it allows efficient spatial averaging due to the large number of uncorrelated speckle spots available. The main advantage of the CCD...
camera as a detector is the access it provides to nonergodic samples and the significantly improved data acquisition time, because a large number of scattering experiments are performed simultaneously. In standard DWS or DLS measurements, the data acquisition time has to be several orders of magnitude larger than the typical relaxation time of the correlation function \( g_2(t) - 1 \), a restriction that does not apply to multispeckle-DWS/DLS (MSDLS, MSDWS). The main drawback of camera-based DLS/DWS is the limited time resolution of CCD cameras. Typically, correlation times \( t \) down to a few ms can be accessed (as compared to 10 ns with a standard photomultiplier-digital correlator setup), which is a particular problem for DWS, where fast relaxation processes are frequently found. However, a combination of two-cell DWS and multispeckle DWS, as shown in Figure 4, turns out to be a perfect combination for overcoming most of the commonly encountered experimental limitations. [50, 59] Using both techniques allows a range of 10 ns to at least 10,000 seconds to be covered (the latter only limited by the waiting time), hence, more than 12 orders of magnitude in correlation time. [50, 53, 57, 59] It is worthwhile to note that it seems impossible to combine the advantages of MSDLS with current multiple scattering suppression schemes, because a well-defined scattering geometry is necessary (as explained before) for obtaining a nonzero cross-correlation signal.

**APPLICATIONS**

In the following examples, we will illustrate how the previously discussed techniques can be applied to the study of fluid and solid-like soft materials.

**Positional Correlations and Dynamics in Suspensions of Charged Spheres**

In the first example (Figures 7 and 8), a 3DDLs measurement of two moderately concentrated colloidal suspensions in a water/ethanol mixture is shown (particle radius \( a = 58.7 \) nm). [30, 60] In this concentration regime, the samples already appear strongly turbid, which makes the application of traditional static light scattering impossible. The 3DDLs allows quantitative information about the mesoscopic interparticle interactions and dynamics in this regime to be obtained. In the classical DLVO theory for charged particles, the (kinetic) stability is explained by assuming that a repulsive Debye–Hückel-type interaction superimposes to the attractive van der Waals interaction, resulting in a net repulsion or attraction depending on the screening by the surrounding electrolyte. [1–3] In a fully deionized suspension, repulsive forces are therefore strong, leading to short-range order (or even crystallization) in the particle positions, as characterized by the radial pair correlation function \( g(r) \).

The particle dynamics in dense colloidal suspensions is influenced by hydrodynamic interactions (HI), particularly if strong repulsive forces are present. [61] HI are mediated by the solvent in which the particles are suspended. They are specific to colloidal suspensions and do not have an analog in atomic physics (such as direct interactions). [7] In dynamic light scattering, Eq. 9, HI can be taken into account by introducing an effective \( q \)-dependent diffusion constant: \( D_{\text{eff}}(q) = D_0 H(q) S(q) \), and \( H(q) \) can be extracted from the measured static and dynamic structure factor. [5, 30]
Excellent agreement between experimental and theoretical calculations is found for concentrations up to about $\Phi \approx 1\%$ (theory: pairwise additivity approximation (PA)\cite{61}). HI in more concentrated systems deviate from this model\cite{30,62}, but a detailed understanding of this behavior is still lacking.

Figure 7. Suspensions of polystyrene latex particles (radius $a = 58.7$ nm) at different volume fractions (from left to right): $\Phi \approx 0.01\%$. Deionized suspensions at $\Phi = 0.13\%$, 0.42\%, and 1.05\% (from left to right).

Figure 8. Left: Static structure factors $S(qa)$ of deionized suspensions of polystyrene latex particles (volume fraction $\Phi$, radius $a = 58.7$ nm) measured with 3DDL5 at 25°C. Solid lines: Best-fit polydisperse HNC calculations using a Yukawa interaction potential with adjustable volume fraction and effective charge $Z_{\text{eff}}$\cite{30,61}. Right: Hydrodynamic function of charged sphere suspensions determined by 3DDL5. Solid lines: Pairwise additivity approximation (PA).
Colloidal Aggregates and Gels

Aggregation and gelation (Figure 9) in complex fluids has been, for a long time, a field of intense research, where fundamental as well as applied questions are equally important. Applications of gels and sol–gel processing include such different areas as ceramics processing, cosmetics and consumer products, food technology, to name only a few. Gels are formed by chemical or physical reactions of small subunits (molecules, polymers, or colloids) that can be reversible or irreversible.

The macroscopic features that bring together such different materials are based on the microstructural properties of all gels, which can be described as random networks built by aggregation of individual subunits. Starting from a solution of the subunits, the system is destabilized, which leads to aggregation, cluster formation, and gelation. At the gel point, a liquid–solid transition is observed that can be characterized by the appearance of a storage modulus in rheological measurements. In dilute colloidal gel networks, the individual clusters show a fractal structure leading to a power law dependence of the structure factor $S(q) \propto q^{-d_f}$ for $1/a < 1/q < R_c$. The fractal dimension is a measure for the compactness of the individual cluster—the higher the $d_f$, the more compact the clusters. For diffusion-limited cluster aggregation (DLCA) $d_f \approx 1.8$, while for reaction-limited cluster aggregation (RLCA), $d_f \approx 2.1$ is expected.[63–68] In the most simple picture, the individual clusters grow at the same rate until they fill the whole accessible volume, with a critical cluster radius given by

$$R_c \approx a \Phi^{-1/(3-d_f)}$$

Under highly dilute conditions, the structure and dynamics of colloidal gels can be analyzed using single-light scattering.[69,70] However, with colloidal suspensions or polymer and biopolymer solutions, the limit of single scattering is quickly reached, and standard SLS and DLS experiments cannot be used to follow aggregation and gelation and to characterize the gel structure. The use of cross-correlation techniques such as 3DDLS can help to expand this regime considerably. This is illustrated with a recent study of the heat-induced aggregation and gelation of the globular protein $\beta$-lactoglobulin.[72] $\beta$-lactoglobulin, the major component of whey, with a molar mass of 18,600 g/mol and a radius of 2 nm, is one of the most intensively studied globular proteins. Globular proteins denature if heated to sufficiently high temperatures, which generally leads to aggregation and the formation of a gel, if the

![Figure 9.](image)

Figure 9. In aggregation and gelation, attractive interparticle interactions lead to the growth of individual (fractal) clusters until they fill the whole accessible volume at $R_c \approx a \Phi^{-1/(3-d_f)}$. 
protein concentration is high enough.\textsuperscript{71} The structure of these gels depends on external conditions such as pH, ionic strength, protein concentration, and temperature. For \(\beta\)-lactoglobulin, the aggregation process can be quenched by rapidly lowering the temperature. At room temperature, the aggregation is very slow, and if the system is diluted, it remains stable, as no further aggregation or break-up of the aggregates is observed for a period of months. It thus becomes possible to characterize the diluted aggregates using scattering techniques at different stages of the aggregation process up to the gel point. However, it is not possible to study more concentrated systems and the gels with standard light scattering techniques, because these systems are too turbid. While neutron and x-ray scattering can be used for turbid systems and applied correspondingly, they probe the gel structure only on length scales smaller than about 50 nm. Using 3DDLs, the effect of multiple scattering on the scattered light intensity can now be effectively corrected, even if the transmission is only 1\%. It was thus possible to determine the structure factor of aqueous solutions of the globular protein \(\beta\)-lactoglobulin as a function of heating time.\textsuperscript{72} The total scattering intensity and the intensity cross-correlation function (\(G_2(t)\)) were determined over a range of scattering angles between 10 and 150 degrees. For the gelled samples, a fraction of the concentration fluctuations relaxes very slowly or is stationary on the time scale of the experiment, i.e., the samples are nonergodic. As the calculation of the singly scattered intensity \(I_1(q)\) using Eq. 8 requires the correct intercept \(\beta_{12}\) for every scattering angle, the samples were slowly rotated during the measurement so that the rotation velocity determines the terminal relaxation of the correlation functions. This guarantees that the correct intercept is obtained, and the correction for multiple scattering is possible even for the non-ergodic gels. The measurements clearly revealed that the structure factor of the aggregated and the gelled proteins can be described by an Ornstein–Zernike equation of the following form:

\[
S(q) = \frac{I(q)}{I(0)} = \frac{1}{1 + (q\zeta)^2}
\]

where \(\zeta\) is the correlation length of the concentration fluctuations in the system. Shown in Figure 10A is the \(q\)-dependence of the scattered intensity corrected for multiple scattering for a solution of \(\beta\)-lactoglobulin with a concentration of 40 mg/mL at different heating times at a temperature of 76\(^\circ\)C. The inset, where the ratio \(\beta_{12}/\beta_{12}^{\text{ideal}}\) of the measured to the ideal intercept as a function of \(q\) is shown for the different individual measurements, demonstrates the effect of multiple scattering on the original scattering data.

It is possible to generate a master curve from the data before and after the gel point by plotting \(S(q)\) vs. \(q\zeta\), which demonstrates that the local structure of the gel fraction is the same as that of the individual aggregates. This is shown in Figure 10B.

However, the use of the 3DDLs technique also has its limits, as measurements for \(\beta\)-lactoglobulin gels were only possible at concentrations below approximately 80 mg/mL. This restriction becomes even more important for colloidal gels, where the scattering cross section of the primary particles is much larger, and the limit of the 3DDLs technique to provide a measurement of the single-scattering \(S(q)\) and \(S(q, t)\) is reached at much lower particle concentrations.
For concentrated colloidal dispersions, DWS is able to provide information about the dynamics during aggregation and gelation. Shown in Figure 11 is the measured autocorrelation function as a function of time after a fluid–solid sol–gel transition. A suspension of monodisperse polystyrene latex spheres is destabilized by increasing the solvent ionic strength with a catalytic reaction. Thereby, the electrostatic repulsion of the double layer is reduced, and the particles aggregate due to van der Waals attraction. At early stages, clusters form due to particle aggregation, and the decay of the correlation function shifts to higher correlation times due to the slower motion of the clusters (data not shown). Gelation occurs when a single cluster fills the entire sample volume. After the sol–gel transition, we observe that the correlation function $g_2(t) / g_2(0)$ does not decay to zero but remains finite. At the gel time, the time dependence of the mean-squared displacement changes qualitatively from diffusion to arrested motion well described by:

$$\langle \Delta r^2(t) \rangle = \delta^2 \left[ 1 - e^{-t/\tau_p} \right]^{p/4}$$  \hspace{1cm} (33)

At short time intervals, this expression reduces to a power law $\Delta r^2(t) \propto \tau_p$, and the exponent for diffusion $p = 1$ drops rapidly at the gel point and takes a value of $p \approx 0.7$.
for all \( t > t_{gel} \). Similar features of the fluid–solid transition have been observed for many colloidal systems, such as in ceramic processing of aluminum oxide powders or in yogurt and cheese making. \( [31,33,73] \)

It is possible to directly link the results from DWS to the macroscopic storage modulus, taking advantage of a recent model developed by Krall and Weitz. Once the gel spans the whole sample, the signal is dominated by a broad distribution of elastic gel modes. For the case of fractal gels, the storage modulus is given by

\[
G_0' = G'(\omega \gg 0) = 6\pi \eta /\tau_c. \tag{70}
\]

**DWS-Based Tracer Microrheology**

The underlying idea of optical microrheology is to study the thermal responses of small (colloidal) particles embedded in a material.\(^74\)–\(^80\) Thereby, it is possible to obtain quantitative information about the loss and storage moduli, \(G'(\omega)\) and \(G''(\omega)\), over an extended range of frequencies.\(^81\),\(^82\) This technique was introduced some years ago when Mason and Weitz suggested a quantitative relation between the tracer mean-squared displacement and the complex shear modulus (Refs. \([77–79]\)).\(^e\)

\[
\tilde{G}(s) = \frac{s}{6\pi a} \left[ \frac{6k_B T}{s^2 \langle \Delta r^2(s) \rangle} \right] \quad (34)
\]

One of the most popular techniques with which to study the thermal motion of the tracer particles is diffusing wave spectroscopy (DWS), because it allows relatively easy

\(^e\)Recently, a rigorous theoretical derivation (albeit certain constraints) for this generalized Stokes–Einstein relation was reported in Refs. \([83,84]\).
access to small particle displacements over an extended range of distances from less than 1 nm to about 50 nm. Thereby, elastic moduli from less than 1 Pa to 1 MPa can be accessed in a two-cell or multispeckle DWS experiment (Refs. [31,85,86]). Shown in Figure 12 are some recent measurements, where tracer particles (polystyrene spheres, diameter 720 nm and 1500 nm) were introduced in an otherwise transparent matrix consisting of a concentrated surfactant solution (aqueous solution of the surfactant molecule hexa-ethylene glycol mono n-hexadecyl ether (C\textsubscript{16}E\textsubscript{6}) at 100 mg/ml. Under the chosen conditions, these surfactants form giant, polymer-like micelles, which results in a surprisingly strong viscoelastic liquid.[59,82,87] Good agreement between classical rheology and DWS-based microrheology is found, with a dramatically increased frequency range for the latter technique. For a quantitative comparison, a scaling factor of 3/2 had to be introduced. The origin of this factor is not yet well understood but likely reflects the imperfect coupling of the tracer sphere to the medium (for details, see Ref. [59]).

**Summary and Outlook**

In the past, light scattering probes have been major tools for physicists and chemists to use to access local nano- and mesoscopic properties of complex fluids.

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1DWS can detect particle displacements in the subnanometer range with visible light. For (the limiting case of) \(L/l^* = 150\), a displacement of \(\sqrt{\langle \Delta r^2(t) \rangle} \approx 1 \text{ nm}\) leads to a detectable decay of \(g_2(t) - 1 \approx 0.99\). For a purely elastic solid material, we find from Eq. 34, \(G = kT/(\pi a^3)\) (with \(\langle \Delta r^2(s) \rangle = \delta^2\) and \(1 = 1/s\)). Hence, a 1 \(\mu\)m tracer particle and a still detectable 0.1 nm displacement corresponds to a measurable elastic modulus of ca. 1 MPa.
With the recent developments summarized in this article, it has now become possible to study a new class of solid-like and turbid materials. Currently, these techniques are mainly used by specialists. However, in the near future, we expect them to spread rapidly in academia and applied research. The first instruments of this generation are already commercially available,[88] and applications in industrial environments are underway.[89]

In this article, we tried to give a comprehensive guideline to the different methods while using a minimum of mathematical expressions. Furthermore, some examples were given on how to perform light scattering on solid-like and turbid materials. Other important systems of this class that remain unmentioned include polymer solutions and gels, colloidal glasses, emulsions, ceramic slurries and green bodies, pastes, and all kinds of soft viscoelastic fluids or solids accessible to DWS-based tracer micro-rheology. In summary, we can say that it is now possible to access any kind of soft material using light scattering techniques, provided the system is homogeneous on macroscopic length scales.

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