

Dynamical properties of multiply scattered light from independent Brownian particles

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We have measured the angle and concentration dependence of the first cumulant of the field-correlation function for multiply scattered light from polystyrene latex particles in water. The experimental results are analyzed in terms of a model based on the assumptions that the scattering particles are pointlike and that the number of renormalized scattering events is Poisson distributed. The agreement between theoretical and experimental results is satisfactory.

I. INTRODUCTION

The recent rapid development of dynamic-light-scattering methods in the study of macromolecular motion has renewed the interest in multiple scattering of light. Since the usual single-scattering assumption breaks down, drastic changes in the dynamic and static properties of the field-correlation (FC) function may occur.

Several authors have analyzed multiple scattering from noninteracting Brownian particles by the use of lowest-order multiple-scattering corrections.¹⁻⁴ Such corrections have also been applied in the analysis of experimental results from optically dense systems.^{4,5}

Multiple scattering has also gained much interest in connection with light scattering from molecular fluids.⁶ Of particular interest to the present work are recent papers by Bedeaux and Mazur,⁷ and by Boots *et al.*^{8,9} These papers show that a renormalization technique can be used in which the pure extinction of the light beam, as it passes through the scattering volume, is taken into account in the definition of the permittivity tensor. In this formalism, multiple scattering is expressed in terms of renormalized "single" scattering, "double" scattering and so forth.

The aim of the present paper is to calculate the first cumulant of the FC function to any order of multiple scattering and compare the resulting expressions with experimental results. The paper extends the first-order analysis developed previously,⁴ using the idea of renormalization.

The paper is organized as follows: In Sec. II we discuss multiple scattering and introduce a diagram representation suitable for discussing various scattering processes in terms of density correlation of the scattering particles. In Sec. III this formalism is applied to scattering from noninteracting Brownian particles, and renormalization resulting from consideration of the time dependence of the various diagrams is also shown. In Sec. IV we develop an explicit expression for

the angle and concentration dependence of the first cumulant of the FC function of light scattered from pointlike isotropic scatterers. This is done for both the polarized and depolarized components, using a Poisson distribution for the number of "renormalized" scattering events.

Our experiments are discussed in Sec. V. The results are compared with calculated values using the expressions developed in Sec. IV.

II. MULTIPLE SCATTERING EXPANSION OF THE FC FUNCTION

The scattered field $\vec{E}(t)$ can be written as a sum of partial fields corresponding to the order of multiple scattering⁴:

$$\vec{E}(t) = \sum_{n=1}^{\infty} \vec{E}_n(t). \quad (1)$$

Consequently, the FC function $G(t)$ of the scattered field is given by

$$G(t) \equiv \langle \vec{E}^*(t) \vec{E}(0) \rangle = \sum_{i,m} G_{i,m}(t), \quad (2)$$

where the partial FC function $G_{i,m}(t)$ is defined by

$$G_{i,m}(t) \equiv \langle \vec{E}_i^*(t) \vec{E}_m(0) \rangle. \quad (3)$$

The explicit form of the n th-order scattered field, and hence the various partial FC functions, depends on factors such as the scattering geometry, the properties of the scattering medium, and so forth. Such explicit expressions have been given in Ref. 4, where it is shown that the fluctuation of the scattered field is connected to the properties of the scattering medium through Fourier components $\psi(\vec{q}, t)$ of the instantaneous particle density $\rho(\vec{r}, t)$:

$$\psi(\vec{q}, t) = V^{-1} \sum_{i=1}^N e^{-i\vec{q} \cdot \vec{r}_i(t)}, \quad (4)$$

where N is the number of scattering particles within the scattering volume V .

Each scattering of the field, involving a momentum transfer \vec{q} , introduces a density component $\psi(\vec{q}, t)$. The fluctuating part of the n th-order scattered field is then given by the product $\prod_{i=1}^n \psi(\vec{q}_i, t)$. Hence, the statistical properties of the partial FC function $G_{l,m}$ depends on the density-correlation functions $D_{l,m}(t)$,

$$D_{l,m}(t) = \left\langle \prod_{i=1}^l \psi^*(\vec{q}_i, t) \prod_{j=1}^m \psi(\vec{q}'_j, 0) \right\rangle. \quad (5)$$

Such density-correlation functions are conveniently discussed using a diagram representation in which each open circle “ i ” represents $\psi^*(\vec{q}_i, t)$ in the upper line and $\psi(\vec{q}'_j, 0)$ in the lower line. The density-correlation function $D_{l,m}(t)$ can then be written

$$D_{l,m}(t) = \begin{array}{c} \circ \cdot \cdot \cdot \circ \\ \circ \cdot \cdot \cdot \circ \\ m \end{array} \quad (6)$$

The correlation range depends, of course, on the system in question, but generally the density-correlation function $D_{l,m}(t)$ can be expressed as the sum of terms containing all possible correlated parts. As an example, we shall consider the density correlation function $D_{2,1}(t)$. From Eqs. (4) and (5) one notes that $D_{2,1}(t)$ contains the summation over three “generic” particles i, j, l which may be grouped in the following manner:

$$i, j, l \rightarrow [i, j, l] + [i], [j, l] + [i, l], [j] + [i, j], l + [i], [j], [l], \quad (7)$$

where the particles inside the square brackets are correlated. The first term on the right-hand side of Eq. (7) corresponds to three correlated particles and the last term to three uncorrelated particles. The diagram representation of the corresponding correlation function $D_{2,1}(t)$ is

$$\begin{array}{c} \circ \circ \\ \circ \end{array} = \begin{array}{c} \circ \text{---} \circ \\ | \\ \circ \end{array} + \begin{array}{c} \circ \text{---} \circ \\ | \\ \circ \end{array} + \begin{array}{c} \circ \text{---} \circ \\ | \\ \circ \end{array} + \begin{array}{c} \circ \text{---} \circ \\ | \\ \circ \end{array} + \begin{array}{c} \circ \text{---} \circ \\ | \\ \circ \end{array} \quad (8)$$

On the right-hand side of Eq. (8) lines are used to symbolize summations over correlated particles and closed circles are used to distinguish diagrams for correlated particles (CP diagrams) from those corresponding to the density-correlation function $D_{2,1}$.

Defining a correlated part of a diagram as either closed circles connected by lines or isolated closed circles, the value of a CP diagram is given by the product of the contributions from each correlated part.

Symbolically, Eq. (8) may be written

$$D_{2,1}(t) = \sum_{\alpha=1}^5 d_{2,1}(\alpha, t), \quad (9)$$

where $d_{2,1}(\alpha, t)$ represents a CP diagram and α is a counting index.

A general diagram $D_{l,m}(t)$ is then given by the sum of all distinct CP diagrams $d_{l,m}(\alpha, t)$,

$$D_{l,m}(t) = \sum_{\alpha} d_{l,m}(\alpha, t). \quad (10)$$

The partial FC function $G_{l,m}(t)$ is obtained from Eq. (10) by multiplying $D_{l,m}(t)$ by an appropriate factor $F_{l,m}(\{\vec{k}\})$, taking into account the scattering geometry, etc., and then summing over all intermediate wave vectors $\{\vec{k}\}$. The general connection between the partial FC function of the scattered field and the statistical properties of the density fluctuations of the scattering medium is then given by

$$G_{l,m}(t) = \sum_{\{\vec{k}\}} \sum_{\alpha} F_{l,m}(\{\vec{k}\}) d_{l,m}(\alpha, \{\vec{k}\}, t). \quad (11)$$

In Sec. III this formalism shall be applied to non-interacting Brownian particles.

III. MULTIPLE SCATTERING FROM NONINTERACTING BROWNIAN PARTICLES

In the case of noninteracting Brownian particles, each particle is only correlated with itself. Hence, the value of a correlated part of a diagram, consisting of n filled circles on the upper line and m in the lower, is simply given by

$$\begin{array}{c} \circ \text{---} \circ \text{---} \circ \\ | \\ \circ \text{---} \circ \text{---} \circ \\ m \end{array} = V^{-(l+m-1)} \rho F_s(\vec{Q}_l, t) \delta_{\vec{Q}_l, \vec{Q}_m}, \quad (12)$$

where ρ is the number density N/V , $\vec{Q}_l = \sum_{i=1}^l \vec{q}_i$ and $\vec{Q}_m = \sum_{j=1}^m \vec{q}'_j$, and $F_s(\vec{Q}, t)$ is the self-intermediate scattering function which, for noninteracting Brownian particles, is given by

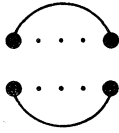
$$F_s(\vec{Q}, t) = e^{-D Q^2 t}, \quad (13)$$

where D is the infinite-dilution diffusion coefficient.

In the following, we assume that the Brownian particle is so small that intraparticle multiple scattering can be ignored. Thus, diagrams containing lines between neighboring circles in either the upper or the lower line are excluded.

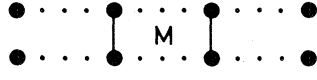
Two useful results follow directly from Eq. (12):

(i) A correlated part containing only circles on one of the lines has vanishing momentum transfer,



$$\propto \rho^2 \delta_{\vec{Q},0} \delta_{\vec{Q}',0}. \quad (14)$$

(ii) All diagrams having M distinct lines connecting the upper and lower line have the same time dependence,



$$\propto \prod_{j=1}^M F_s(\vec{q}_j, t). \quad (15)$$

This leads to the conclusion that the time dependence of a diagram is determined by the number of lines connecting the upper and lower line. All other properties of the diagram influence its value, but not its time dependence. Thus, a diagram $D_{l,m}(t)$ may alternatively be expressed as a sum of diagrams having 1 up to $M = \min(l, m)$ lines connecting the upper and lower line:

$$D_{l,m}(t) = \sum_{n=1}^M D_{l,m}(n, t), \quad (16)$$

where

$$D_{l,m}(n, t) = \sum_{\alpha}^n d_{l,m}(\alpha, t). \quad (17)$$

The summation \sum_{α}^n in Eq. (17) contains all CP diagrams $d_{l,m}(\alpha, t)$ having n lines connecting the upper and lower line.

Using Eq. (15), the diagram $D_{l,m}(n, t)$ can be expressed as

$$D_{l,m}(n, t) = D_{l,m}(n, 0) \prod_{j=1}^n F_s(\vec{q}_j, t). \quad (18)$$

In the same way we may also define a partial FC function, $G_{l,m}(n, t)$,

$$G_{l,m}(n, t) = \sum_{\{\vec{k}\}} F_{l,m}(\{\vec{k}\}) D_{l,m}(n, \{\vec{k}\}, 0) \times \prod_{j=1}^n F_s(\vec{q}_j, t). \quad (19)$$

When summing $G_{l,m}(t)$ over l and m , we group together all the partial FC functions $G_{l,m}(n, t)$ having n lines connecting the upper and lower line. Thus we define the n th-order partial FC function $G_n(t)$ by

$$G_n(t) = \sum_{l,m}^n G_{l,m}(n, t). \quad (20)$$

This leads to the following renormalized expres-

sion for the normalized FC function $g(t)$ of the scattered field:

$$g(t) = \frac{G(t)}{G(0)} = \sum_{n=1}^{\infty} g_n \left\langle \prod_{j=1}^n F_s(\vec{q}_j, t) \right\rangle_n, \quad (21)$$

where $g_n = G_1(0)/G(0)$ has the form of a probability density, and the average value appearing in Eq. (21) is defined as

$$\begin{aligned} & \left\langle \prod_{j=1}^n F_s(\vec{q}_j, t) \right\rangle_n \\ &= \left(\sum_{l,m}^n \sum_{\{\vec{k}\}} F_{l,m}(\{\vec{k}\}) D_{l,m}(n, \{\vec{k}\}, 0) \right)^{-1} \\ & \times \left(\sum_{l,m}^n \sum_{\{\vec{k}\}} F_{l,m}(\{\vec{k}\}) D_{l,m}(n, \{\vec{k}\}, 0) \prod_{j=1}^n F_s(\vec{q}_j, t) \right). \end{aligned} \quad (22)$$

Each term in the summation on the right-hand side of Eq. (21) represents the renormalized n th-order scattering with the extinction included in the factor $g_n(0)$.

One feature of the procedure outlined above is that the resulting equations, Eqs. (21) and (22), can be applied for model calculations by which essential dynamical properties of multiple scattering can be discussed.

IV. EVALUATION OF THE FIRST CUMULANTS USING A POISSON DISTRIBUTION

As a first approximation we assume the number of "renormalized scattering events" to be Poisson distributed. This corresponds to independent scattering events, each having the same probability. The first cumulant of the FC function is given as¹⁰

$$\begin{aligned} K_1 &= \lim_{t \rightarrow 0} \left(-\frac{\partial}{\partial t} \ln g(t) \right) = \sum_{n=1}^{\infty} g_n \sum_{j=1}^n \langle Dq_j^2 \rangle_n \\ &= \sum_{n=1}^{\infty} g_n K_{1,n}. \end{aligned} \quad (23)$$

In Eq. (23) the first cumulant K_1 is expressed as a weighted sum of partial first cumulants $K_{1,n}$ corresponding to the renormalized n th-order scattered light. The partial first cumulant is, in turn, a sum of n average single-scattering decay constants. If, as pointed out by Sorensen *et al.*,³ we restrict ourselves to pointlike particles, we may use a symmetry argument. The first cumulant corresponding to double scattering $K_{1,2}$ is then angle independent and equal to that obtained for single scattering at 180°. The double-scattering process may be visualized as two independent

scattering events. Due to the fact that pointlike particles are isotropic scatterers, the average scattering angle for each scattering is 90° . Two scatterings at 90° yield on the average an effective 180° scattering process, which explains the initial slope (first cumulant) of the double-scattering FC function.

Applying these arguments to an arbitrary n th-order ($n > 2$) multiple-scattering process the first cumulant [Eq. (23)] may be written

$$K_1(\theta) = \langle n \rangle \Gamma(90^\circ) - g_1 [\Gamma(90^\circ) - \Gamma(\theta)], \quad (24)$$

where θ is the angular position of the detector, $\Gamma(\theta)$ is the single-scattering decay constant, and $\langle n \rangle$ is the average order of multiple scattering. Thus, for independent pointlike particles, the value of the first cumulant K_1 at $\theta = 90^\circ$ is equal to that of the average number $\langle n \rangle$ of multiple scattering.

For a more detailed analysis of the angle- and concentration dependence of K_1 , the probability distribution g_n must be used. We then distinguish between the polarized component K_{1p} , which includes single scattering as well as multiple scattering, and the depolarized component K_{1n} , which does not contain single scattering. Using the Poisson distribution for g_n , we obtain

$$K_{1p}(\theta) = (1 - e^{-\langle n \rangle_p})^{-1} \langle n \rangle_p \Gamma \{ [1 - [1 - \gamma(\theta)] e^{-\langle n \rangle_p}] \} \quad (25)$$

and

$$K_{1n}(\theta) = [1 - (1 + \langle n \rangle_n) e^{-\langle n \rangle_n}]^{-1} \langle n \rangle_n \times \Gamma(1 - e^{-\langle n \rangle_n}). \quad (26)$$

Here, $\gamma(\theta) = \Gamma(\theta)/\Gamma(90^\circ)$, $\langle n \rangle_p$ is the average number of scattering events in the polarized component, $\langle n \rangle_n$ is the average number of scattering events in the depolarized component, and the factors $(1 - e^{-\langle n \rangle_p})^{-1}$ and $[1 - (1 + \langle n \rangle_n) e^{-\langle n \rangle_n}]^{-1}$ are due to normalization of g_n .

Finally, we add that for a Poisson distribution the average number of scattering events is proportional to the concentration C of scattering particles:

$$\langle n \rangle_p = \alpha_p C, \quad (27a)$$

$$\langle n \rangle_n = \alpha_n C. \quad (27b)$$

V. EXPERIMENTAL RESULTS AND DISCUSSION

The spectrometer used consisted of an Ar⁺-ion laser ($\lambda = 514.5$ nm, 1.4 W), a movable ITT-FW 130 photomultiplier (PM) tube and a 24-channel Malvern digital correlator. Rectangular sample cells of 1 cm optical path were placed in a water bath for index matching.

The scattering systems consisted of filtered

water suspensions of polystyrene latex spheres obtained from Dow Chemical Co. The average diameter of these spheres was reported by the manufacturer to be 109 nm with a standard deviation of 2.5%. Light scattering at a concentration $C = 5 \times 10^{-7}$ g/cm³ gave a hydrodynamic diameter of 115 nm and this value was used in the calculations. The small polydispersity is not expected to seriously distort the measurements of the FC function.

A lens was used to focus the laser beam down to about 200–300 μ m, which is approximately in the size of the scattering volume as defined by the apertures in front of the PM tube. The reason for the lens was to obtain a higher intensity of the depolarized scattered light, especially at the lowest concentrations. But, as pointed out by Sorensen *et al.*³ this tends to give a too large first cumulant for the polarized component since multiply scattered light is collected from a larger volume than singly scattered light. This effect is expected to influence our measurements, especially at low concentrations. In addition, the use of a focusing lens introduces an uncertainty due to the possibility that the scattering volume as determined by the apertures does not fully coincide with that determined by the focused beam. To reduce this uncertainty, we adjusted the lens to maximum intensity in every run. A better approach would have been to use a smaller sample cell and illuminate it as uniformly as possible.

We used Glan polarizers with an extinction coefficient of about 10^{-5} to determine the polarization of the incident light and to single out the polarized and depolarized components of the scattered light. In the measurements of the depolarized component, the analyzing polarizer was adjusted to minimum intensity. However, an extinction not better than 10^{-5} inevitably leads to some mixing of the slower fluctuating polarized component into the depolarized one, especially at the lowest concentrations. This tends to give a too small value for the first cumulant of the depolarized light at these concentrations.

The experimental data were fed into a computer programmed to find the initial slope of the normalized FC function. In experiments on multiple scattering it is particularly important to use a correct sample time on the correlator. The reason is that the scattered light is a sum of components which can have widely different fluctuation times. Hence, the rule of thumb to select a sample time $T \approx 0.1T_c$, where T_c is the average correlation time is incorrect since this results in an integration of the fastest components. An example of such an effect is given in Fig. 1, which shows that the value of the first cumulant obtained at $T \approx 0.1T_c$ is too

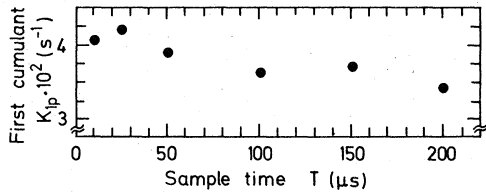


FIG. 1. Experimentally measured first cumulant K_{1p} vs sample time T on correlator. Scattering angle was 30° and $T_c \approx 2.5$ ms.

low by about 25%.

The analysis of our experimental data is based on the Siegert relation which connects the intensity-correlation function to the FC function.¹¹ This relation requires Gaussian statistics, which are not exactly fulfilled in the case of multiple scattering. However, as pointed out by Kelly,² the non-Gaussian component is negligibly small, except at very high degrees of multiple scattering. Furthermore, Colby *et al.* did not find any significant contribution from a non-Gaussian component in their experiments on dense optical systems.⁵

In Figs. 2, 3, 4, and 5 the experimental results are compared with calculated values using Eqs. (25) and (26). In Fig. 2, values for the first cumulant of the polarized component K_{1p} are plotted as a function of concentration. The full line is calculated from Eq. (25) using the value $\alpha_p = 5.4 \times 10^2$ cm³/g.

The fit between the theoretical curves and the experimental values is quite good in the concentration range studied. At low concentration, the

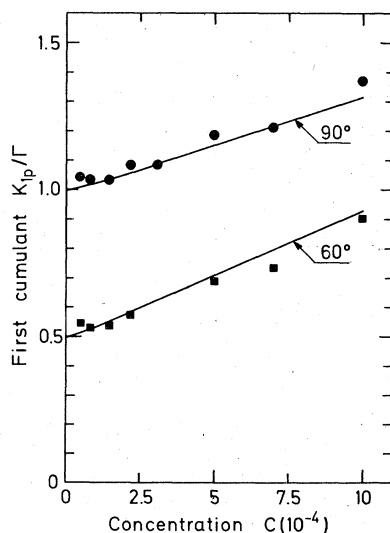


FIG. 2. Experimentally measured first cumulant K_{1p}/Γ vs concentration at scattering angles $\theta = 90^\circ$ and $\theta = 60^\circ$. The full curve is calculated from Eq. (25) in the text.

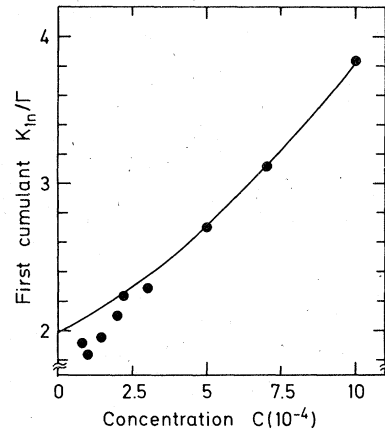


FIG. 3. Experimentally measured first cumulant K_{1n}/Γ vs concentration at a scattering angle $\theta = 90^\circ$. The full curve is calculated from Eq. (26).

experimental values are too high, which may be due to the use of a focusing lens.

Figure 3 shows the first cumulant of the depolarized component K_{1n} as a function of concentration. Here the discrepancy at low concentration is probably due to the following two factors: (i) the latex particles used are too large to be treated as point particles, hence the "average scattering angle" is less than 90° ; (ii) the mixing of the slower polarized component with the depolarized one leads to a too small first cumulant. The full line in Fig. 3 is calculated from Eq. (26) using the value $\alpha_n = 3.3 \times 10^3$ cm³/g.

Comparing the values for α_p and α_n , we find that the average amount of multiple scattering in the depolarized component is about six times higher

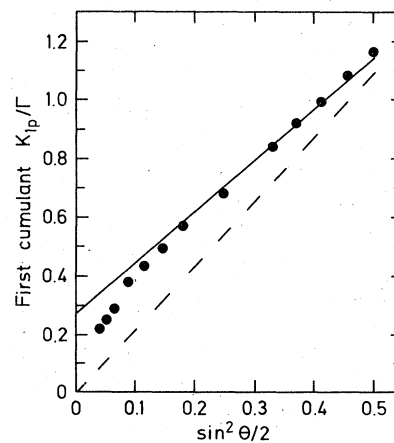


FIG. 4. Experimentally measured first cumulant K_{1p}/Γ vs angle parameter $\sin^2 \frac{1}{2} \theta$ at a concentration $C = 5 \times 10^{-4}$ g/cm³. The full curve is calculated from Eq. (25), the broken curve is the expected single-scattering curve.

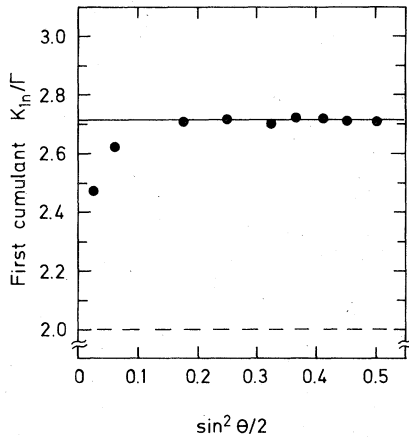


FIG. 5. Experimentally measured first cumulant K_{1n}/Γ vs angle parameter $\sin^2 \theta/2$ at a concentration $C = 5 \times 10^{-4}$ g/cm. The full curve is calculated from Eq. (26), the broken curve is the expected double-scattering curve:

than that in the polarized one. Our calculation provides no quantitative explanation for this result. Qualitatively, this effect is not surprising since the incident polarized light is gradually depolarized through multiple scattering. Hence, contributions from higher-order multiple scattering are relatively more important in the depolarized than in the polarized component.

In Fig. 4, we have plotted the first cumulant of the polarized component as a function of scattering angle at a concentration of about 5×10^{-4} g/cm³. Equation (25) gives a linear relationship between the first cumulant K_{1p}/Γ and the angle parameter $x = \sin^2 \theta/2$. The intercept at $x = 0$ corresponds to the average number $\langle n \rangle_p$ of scatterings. The full curve in Fig. 4 is calculated from Eq. (25) with the value for α_p used in Fig. 2. The broken curve represents single scattering. Except at small angles, the agreement between theoretical and experimental values is satisfactory. The discrepancy is probably due to dust in the sample and to the fact that the latex particles are not pointlike.

Similarly, Eq. 26 predicts the first cumulant K_{1n}/Γ of the depolarized component to be angle independent. This is shown in Fig. 5, using the value for α_n obtained from Fig. 3. Again, the agreement between the theoretical predictions and the experimental values is good except at small angles.

The two main sources for this discrepancy are probably: (i) the polarized component mixed into the depolarized one becomes slower the smaller the angle and (ii) depolarized scattering from dust

and the cuvette is usually more pronounced for small angles.

VI. CONCLUSION

The diagram technique developed in the present paper is suitable for analyzing various multiple-scattering processes in terms of particle-density correlations. When applied to a scattering system of noninteracting Brownian particles, this technique results in a renormalization procedure. The essential point is that many of the various scattering processes have the same time dependence, thus formally the renormalization is carried out by adding all diagrams having the same time dependence. The FC function of the scattered field is then given as a sum of renormalized n th-order partial FC functions. Qualitatively, the renormalization takes care of the extinction of the n th-order scattered light.

The renormalized expression for the FC function leads to a simple expression for the first cumulant. Assuming isotropy, which is valid for pointlike particles, the first cumulant is a direct measure of the average number of multiple-scattering events. Assuming further that the number of renormalized scattering events has a Poisson distribution, we obtain simple expressions for the angle and concentration dependence of the first cumulants containing the average number of scatterings as the only parameter. The agreement between values calculated from these expressions and experimental ones is satisfactory.

The final expressions for the first cumulant are restricted to independent pointlike particles and scattering events obeying Poisson statistics. The effect of particle interactions is ignored since they are not expected to be important in our experiments.⁵ We believe a similar renormalization procedure may be useful in even more complicated systems of correlated particles as long as the correlation range is reasonably small compared with the dimension of the scattering volume.

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