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Role of multiple scattering in cross-correlated light scattering with a single laser beam

James A. Lock

Previous systems for measuring cross-correlated light scattering by small particles suspended in a liquid with multiple-scattering suppression have illuminated the particles with two laser beams. It is shown that multiple-scattering suppression should also occur in cross correlation for a system that employs a single laser beam and two closely spaced detectors with wide fields of view. The single-scattering, double-scattering, and single-double-scattering cross-term contributions to the intensity cross-correlation function are calculated. It is found that the two cross terms, when added together, are unimportant for both autocorrelation and cross correlation. The amplitude of the double-scattering term can be greatly diminished by judicious detector spacing because the spatial coherence area in the detector plane for double scattering is much smaller than that for single scattering. © 1997 Optical Society of America

1. Introduction

Over the past three decades, dynamic light scattering has become the nonintrusive measurement technique of choice for studying small particles suspended in a liquid. Single scattering dominates if the suspension is dilute. In this case the autocorrelation function of the light scattered by noninteracting particles decays exponentially with time, and the particle diffusion coefficient is proportional to the decay rate.¹ Multiple scattering is dominant in dense suspensions, and its presence complicates the time dependence of the autocorrelation function. This complication has been addressed in three ways. For semidilute systems in which multiple scattering first becomes significant, various investigators have calculated and compensated for the effects of double²⁻⁵ and multiple^{6,7} scattering. For dense suspensions in the deep multiple-scattering regime, others have modeled multiple scattering by using photon diffusion⁸⁻¹¹ and radiative transfer^{12,13} methods. Last, others have employed cross correlation by using two laser beams to suppress the contribution of multiple scattering to the cross-correlation function relative to that of single scattering.¹⁴⁻²⁰

In 1981, Phillies proposed a scattering geometry

that uses two counterpropagating laser beams that have the same wavelength and two oppositely positioned detectors, and he predicted that single scattering would be strongly cross correlated whereas multiple scattering would be uncorrelated.¹⁴ Subsequent experiments verified that the expected multiple-scattering suppression occurred.^{15,16} A few years later, the complete cross-correlation function for the two-beam system was calculated,¹⁷ and a refinement that uses two laser beams of different wavelengths with filters in front of the detectors was proposed in order to separate the contributions to the cross-correlation function from scattering at angles θ and $\pi-\theta$. Again, subsequent experiments verified that the expected multiple-scattering suppression occurred for the two-wavelength cross-correlation system.¹⁸⁻²⁰

A common concern for these cross-correlation systems is that they are difficult to align. Thus there has been motivation to find simpler scattering geometries that suppress multiple scattering in cross correlation. The use of monomode optical fibers as detectors²¹⁻²⁵ has opened up new possibilities in this regard, and an autocorrelation backscatter probe operating in the scattering near zone that suppresses multiple scattering has been developed.²⁶

Here I present a theoretical analysis of a recently proposed²⁷ cross-correlation geometry that has been found to suppress multiple scattering in dense suspensions and that is easier to align than the two-beam cross-correlation systems. The system that we study here consists of a single laser beam and two closely spaced detectors with wide fields of view.²⁷⁻²⁹ It is shown that multiply scattered light should cease

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being cross correlated for small detector spacings, whereas singly scattered light ceases being cross correlated for larger detector spacings. Thus if two monomode optical fibers coupled to detectors are positioned beyond the multiple-scattering threshold separation but lie within the single-scattering threshold separation, the multiple-scattering contribution to the cross-correlation function should be strongly suppressed with respect to the single-scattering contribution. A preliminary version of this calculation that ignores both the polarization and the angular dependence of the scattered light has already appeared.³⁰

There are many geometries for which multiple scattering is suppressed in autocorrelation or cross correlation, and the method examined here is only one of them. Another possibility that has been suggested informally by a number of researchers is to perform autocorrelation with the field of view of a collimated detector centered on a point in the laser beam in the scattering cell. At this location the singly scattered light is bright while the multiply scattered light along the detector field of view is much dimmer, again producing multiple-scattering suppression relative to single scattering. Further experimentation will determine which of the many feasible techniques is most efficient at suppressing multiple scattering and which is the easiest to use. The purpose here is more modest, namely, to provide a theoretical analysis of the one-beam cross-correlation technique.

The body of this paper is organized as follows. In Section 2 the scattering geometry is described and the notation is defined. In Sections 3 and 4 single and double scattering, respectively, by a collection of noninteracting spherical particles suspended in a liquid are considered. In Section 5 the two electric-field correlation functions are calculated for the one-beam cross-correlation system. The dominant cross-correlation function includes geometric factors related to the far-zone spatial coherence areas for single and double scattering. In Section 6 the intensity cross-correlation function is calculated, the degree of double-scattering suppression is determined, and the time dependences of the autocorrelation and cross-correlation functions are determined. Finally, in Section 7, results are discussed. In suspensions for which a substantial amount of double scattering occurs, higher orders of multiple scattering are usually important as well. Here, the calculations are limited to double scattering, however, because the spatial coherence area argument for double-scattering suppression can be straightforwardly generalized to higher orders of multiple scattering.

2. Scattering Geometry

We follow the general procedure used to analyze two-beam cross-correlation systems in Refs. 17 and 20. The scattering geometry is illustrated in Fig. 1. A cell of volume V and with transparent walls contains N monodisperse nonabsorbing dielectric spherical particles of real refractive index n_p and radius a suspended in a nonabsorbing liquid of real refractive index n_L .

Fig. 1. Focused laser beam B that has the wave vector $\mathbf{k}_{\text{inc}}$ propagates through a scattering cell of volume V , which contains N particles suspended in a liquid. The field of view of detector d is given by the dashed lines. The origin of coordinates O is at the center of the intersection volume of the beam and the detector field of view.

The average number density of the suspended particles is $\rho = N/V$, their volume fraction is $\phi = 4\pi a^3 \rho / 3$, and their refractive index relative to the liquid is $n = n_p / n_L$. A monochromatic laser beam of wavelength λ in vacuum and angular frequency ω propagates through the cell and is scattered by the particles. The electric field of the unscattered portion of the beam at the position $\mathbf{r}_j$ in the cell at time t_j is

$$\mathbf{E}_{\text{inc}}(\mathbf{r}_j, t_j) = E_0 B(\mathbf{r}_j) \exp(-L_{bj}/2L_{\text{scatt}}) \hat{u}_{\text{inc}} \times \exp(i\mathbf{k}_{\text{inc}} \cdot \mathbf{r}_j - i\omega t_j), \quad (1)$$

where E_0 is the incident field strength, $B(\mathbf{r}_j)$ is a dimensionless function describing the transverse beam profile, L_{bj} is the distance the beam has traveled through the suspension to the position $\mathbf{r}_j$, L_{scatt} is the single-scattering mean free path,³¹ $\hat{u}_{\text{inc}}$ is the beam-polarization direction, and the wave number of the light in the liquid is

$$k \equiv |\mathbf{k}_{\text{inc}}| = \frac{2\pi n_L}{\lambda}. \quad (2)$$

The beam is scattered by particle j at position $\mathbf{r}_j$ at time t_j . The scattered electric field at the far-zone position $\mathbf{R}$ in the cell at later time T is

$$\begin{aligned} \mathbf{E}_{\text{scatt}}(\mathbf{R}, T) = & \frac{E_0 B(\mathbf{r}_j) \mathbf{A}_{\text{Mie}}}{kR} \\ & \times \exp[-(L_{bj} + L_{jR})/2L_{\text{scatt}}] \\ & \times \exp[ikR - i\omega T - i\mathbf{k} \cdot \mathbf{r}_j(t_j)], \end{aligned} \quad (3)$$

Fig. 2. Laser beam B is focused by lens L to half-width R_y . It propagates through a test tube of radius R_z , which contains the liquid and the suspended particles. The test tube is enclosed in a coaxial index-matching vat of radius R_v . Wide-field-of-view detectors α and β with completely overlapping fields of view in the scattering cell are at a distance R_d from the center of the vat.

where the distance in the suspension from particle j to position $\mathbf{R}$ is L_{jR} , the scattered wave vector $\mathbf{k}_{\text{scatt}}$ is

$$\mathbf{k}_{\text{scatt}} = \frac{k\mathbf{R}}{R}, \quad (4)$$

the scattered momentum transfer $\boldsymbol{\kappa}$ is

$$\begin{aligned} \boldsymbol{\kappa} &= \mathbf{k}_{\text{scatt}} - \mathbf{k}_{\text{inc}}, \\ |\boldsymbol{\kappa}| &= 2k \sin(\theta/2), \end{aligned} \quad (5)$$

θ is the scattering angle, and $\mathbf{A}_{\text{Mie}}$ is the far-zone Lorenz–Mie scattering amplitude. For the special case of Rayleigh scattering,³² i.e., $ka \ll 1$, we have

$$\begin{aligned} \lim_{ka \ll 1} \mathbf{A}_{\text{Mie}} &= \mathbf{A}_{\text{Rayleigh}} \\ &= \left(\frac{n^2 - 1}{n^2 + 2} \right) (ka)^3 \hat{\mathbf{k}}_{\text{scatt}} \times (\hat{\mathbf{u}}_{\text{inc}} \times \hat{\mathbf{k}}_{\text{scatt}}). \end{aligned} \quad (6)$$

We consider the case in which the wave vector of the incident laser beam is in the positive z direction. We also assume that the scattering cell is a thin-wall cylindrical test tube with its symmetry axis along the y axis, as shown in Fig. 2. It is surrounded by a coaxial cylindrical vat filled with the same liquid as is in the test tube, corresponding to the experimental geometry of Refs. 27–29. A single-point detector d is placed at the position $\mathbf{R} = \mathbf{R}_d$ in the xz plane for autocorrelation, or a pair of them, α and β , are placed slightly above and below the xz plane for cross correlation, so that

$$\hat{\mathbf{k}}_{\text{scatt}} \approx \sin \theta \hat{\mathbf{u}}_x + \cos \theta \hat{\mathbf{u}}_z. \quad (7)$$

The four input–output polarization channels for this geometry are denoted by VV, VH, HV, and HH and are illustrated in Fig. 3. For single scattering in the xz plane, we can rewrite Eq. (6) for all four polarization channels by using the compact notation

$$\langle \pi_f | \mathbf{A}_{\text{Rayleigh}} | \pi_i \rangle = A_1 [P_1], \quad (8)$$

Fig. 3. Four input–output polarization states for scattering in the xz plane: (a) VV, (b) VH, (c) HV, (d) HH.

where π_i is the initial polarization, π_f is the final polarization (each V or H),

$$A_1 = (ka)^3 (n^2 - 1) / (n^2 + 2), \quad (9)$$

$$[P_1] = \begin{bmatrix} VV_1 & VH_1 \\ HV_1 & HH_1 \end{bmatrix}. \quad (10)$$

In Eqs. (8)–(10) the subscript 1 denotes single scattering. The angular dependence of the elements of $[P_1]$ is

$$\begin{aligned} VV_1 &= 1, \\ VH_1 &= HV_1 = 0, \\ HH_1 &= \cos \theta. \end{aligned} \quad (11)$$

For double scattering by particles j and then l , the scattered field of particle j is the incident field for particle l . Iterating Eq. (6), assuming that l is in the far zone of j , we can write the Rayleigh amplitude for double scattering for all four polarization channels with the detector in the xz plane by using the compact notation $A_2[P_2]$, where

$$A_2 = A_1^2, \quad (12)$$

$$[P_2] = \begin{bmatrix} VV_2 & VH_2 \\ HV_2 & HH_2 \end{bmatrix}, \quad (13)$$

with

$$\begin{aligned} VV_2 &= 1 - \sin^2 \theta_{jl} \sin^2 \phi_{jl}, \\ VH_2 &= \sin^2 \theta_{jl} \sin \phi_{jl} \cos \phi_{jl} \cos \theta \\ &\quad - \sin \theta_{jl} \cos \theta_{jl} \sin \phi_{jl} \sin \theta, \\ HV_2 &= -\sin^2 \theta_{jl} \sin \phi_{jl} \cos \phi_{jl}, \\ HH_2 &= \cos \theta - \sin^2 \theta_{jl} \cos^2 \phi_{jl} \cos \theta \\ &\quad + \sin \theta_{jl} \cos \theta_{jl} \cos \phi_{jl} \sin \theta. \end{aligned} \quad (14)$$

The intermediate wave vector $\mathbf{k}_{jl}$ of the light scattered by j and incident upon l is

$$\mathbf{k}_{jl} = k(\sin \theta_{jl} \cos \phi_{jl} \hat{u}_x + \sin \theta_{jl} \sin \phi_{jl} \hat{u}_y + \cos \theta_{jl} \hat{u}_z). \quad (15)$$

For the remainder of this paper, all quantities in brackets are to be evaluated for all four input–output polarization channels.

Finally, the relative sensitivity¹⁷ of detector d to light that has its last interaction with a particle at position $\mathbf{r}$ is $D(\mathbf{r})$. Only some of the particles in the cell are illuminated by the laser beam and only some of these in turn are in the field of view of the detector. The volume of the laser beam within the cell, the volume of the detector field of view within the cell, and the intersection of these two volumes are denoted by V_1 , V_2 , and V_0 , respectively; the average numbers of particles in these volumes are N_1 , N_2 , and N_0 . The origin of the coordinates is taken to be at the center of V_0 , as in Fig. 1.

3. Single Scattering by a Collection of Particles

We now consider light that reaches detector d at time T because of single scattering of the laser beam by the suspended particles at positions $\mathbf{r}_j$ in the cell at earlier times t_j , as is illustrated in Fig. 4(a). Because the participating particles are both illuminated by the laser beam and in the detector field of view, the single-scattered electric field at the detector is

$$\begin{aligned} [E_{d1}(\mathbf{R}_d, T)] &= \frac{E_0 A_1 [P_1]}{k R_d} \exp(ikR_d - i\omega T) \sum_{j=1}^{N_0} B(\mathbf{r}_j) D(\mathbf{r}_j) \\ &\times \exp[-(L_{bj} + L_{jd})/2L_{\text{scatt}}] \\ &\times \exp[-i\mathbf{k} \cdot \mathbf{r}_j(t_j)], \end{aligned} \quad (16)$$

where the subscripts d and 1 label the detector and denote single scattering, respectively. In obtaining Eq. (16) we assume that the incident laser beam was focused by a lens upstream from the cell, as in Fig. 2, so that the cell lies entirely within the beam focal waist. As a result, $\mathbf{k}_{\text{inc}}$ is independent of $\mathbf{r}_j$. We also assume that the point detector for autocorrelation or the pair of point detectors for cross correlation are placed along the x axis and have wide fields of view, i.e., the radius of a detector field of view in the plane containing the laser beam is much wider than the beam radius. Such detectors have not often been considered because of concerns that they record a large range of $\mathbf{k}$ values, causing instrumental broadening. Only a single-scattered momentum transfer is recorded, and instrumental broadening is thereby avoided, however, if either bare-end monomode optical fibers coupled to detectors are placed on the paraxial focal line of the index-matching vat^{27,28} or if gradient-index lenses are cemented onto the end of the monomode fibers placed far from the vat.²⁹ For these two geometries, all singly scattered light reaching the detectors has the same scattering angle (i.e., $\theta = 90^\circ$) and thus the same scattered momentum transfer.

The noninteracting suspended particles in the scat-

Fig. 4. Single scattering by particle j (a) at time t_j when the particle is at position $\mathbf{r}_j$, (b) at time τ later when the particle has moved by means of diffusion to somewhere within a sphere of radius $\Delta_{\text{ave}}(\tau)$ centered on $\mathbf{r}_j$.

tering cell are in random motion. The probability that particle j is at position

$$\mathbf{r}_j(t_j + \tau) = \mathbf{r}_j(t_j) + \Delta_j(\tau) \quad (17)$$

at time $t_j + \tau$, given that it was at position $\mathbf{r}_j$ at the earlier time t_j , is $P(\Delta_j, \tau)$. The singly scattered electric field reaching the detector at $T + \tau$ averaged over all the possible particle displacements Δ_j at $t_j + \tau$, as in Fig. 4(b), is then

$$\begin{aligned} [E_{d1}(\mathbf{R}_d, T + \tau)] &= \frac{E_0 A_1 [P_1]}{k R_d} \\ &\times \exp[ikR - i\omega(T + \tau)] S^*(\mathbf{k}, \tau) \\ &\times \sum_{j=1}^{N_0} B(\mathbf{r}_j) D(\mathbf{r}_j) \\ &\times \exp[-(L_{bj} + L_{jd})/2L_{\text{scatt}}] \\ &\times \exp[-i\mathbf{k} \cdot \mathbf{r}_j(t_j)]. \end{aligned} \quad (18)$$

In Eq. (18) we assume that the same N_0 particles are within V_0 at times t_j and $t_j + \tau$ and that both the beam profile and the detector field of view are sufficiently slowly spatially varying and τ is sufficiently small so that

$$\begin{aligned} B[\mathbf{r}_j(t_j)] &\approx B[\mathbf{r}_j(t_j + \tau)], \\ D[\mathbf{r}_j(t_j)] &\approx D[\mathbf{r}_j(t_j + \tau)]. \end{aligned} \quad (19)$$

The structure factor of the particles' random motion is

$$S(\mathbf{k}, \tau) \equiv \int d^3\Delta_j P(\Delta_j, \tau) \exp(i\mathbf{k} \cdot \Delta_j). \quad (20)$$

4. Double Scattering by a Collection of Particles

We consider the double-scattering situation of Fig. 5(a). Light scatters from particle j at position $\mathbf{r}_j$ at time t_j and travels the distance

$$r_{jl} = |\mathbf{r}_j(t_j) - \mathbf{r}_l(t_l)| \quad (21)$$

without further interaction to position $\mathbf{r}_l$ at time t_l , where it scatters from particle l . The doubly scattered light then propagates to the detector in the far zone without further interaction with the particles. For this situation, particle j is illuminated by the incident beam, and particle l is in the field of view of the detector. We assume that particle l is also in the scattering far zone of particle j . The far-zone assumption is reasonable for semidilute suspensions because the largest near-zone contribution to the scattered electric field [i.e., the term proportional to $(kr_{jl})^{-2}$] for Rayleigh scattering with^{27,28} $a = 0.053 \mu\text{m}$ is less than 0.1 of the far-zone contribution [i.e., the term proportional to $(kr_{jl})^{-1}$] for $\phi \leq 0.01$. The doubly scattered electric field reaching the detector at time T is then

$$\begin{aligned} [E_{2d}(\mathbf{R}_d, T)] &= \frac{E_0 A_2}{kR_d} \exp(ikR_d - i\omega T) \\ &\times \sum_{j=1}^{N_1} \sum_{l=1}^{N_2} [P_2] \frac{B(\mathbf{r}_j) D(\mathbf{r}_l)}{kr_{jl}} \\ &\times \exp[-(L_{bj} + r_{jl} + L_{ld})/2L_{\text{scatt}}] \\ &\times \exp[i(\mathbf{k}_{\text{inc}} - \mathbf{k}_{jl}) \cdot \mathbf{r}_j(t_j)] \\ &\times \exp[i(\mathbf{k}_{jl} - \mathbf{k}_{\text{scatt}}) \cdot \mathbf{r}_l(t_l)]. \end{aligned} \quad (22)$$

For light reaching the detector at later time $T + \tau$, as in Fig. 5(b), we further assume that $\Delta_j, \Delta_l \ll r_{jl}$ so that the intermediate wave vector from any of the possible positions of the first scattering to any of the possible positions of the second scattering is independent of Δ_j and Δ_l . The doubly scattered electric field reaching the detector at $T + \tau$ averaged over all the

Fig. 5. Double scattering by particles j and l (a) at times t_j and t_l when the particles are at the positions $\mathbf{r}_j$ and $\mathbf{r}_l$, (b) at time τ later when the particles have moved by means of diffusion to somewhere within spheres of radii $\Delta_{\text{ave}}(\tau)$ centered on $\mathbf{r}_j$ and $\mathbf{r}_l$.

possible particle displacements Δ_j and Δ_l at $t_j + \tau$ and $t_l + \tau$ is then

$$\begin{aligned} [E_{2d}(\mathbf{R}_d, T + \tau)] &= \frac{E_0 A_2}{kR_d} \exp[ikR_d - i\omega(T + \tau)] \\ &\times \sum_{j=1}^{N_1} \sum_{l=1}^{N_2} [P_2] \frac{B(\mathbf{r}_j) D(\mathbf{r}_l)}{kr_{jl}} \\ &\times \exp[-(L_{bj} + r_{jl} + L_{ld})/2L_{\text{scatt}}] \\ &\times \exp[i(\mathbf{k}_{\text{inc}} - \mathbf{k}_{jl}) \cdot \mathbf{r}_j(t_j)] \\ &\times \exp[i(\mathbf{k}_{jl} - \mathbf{k}_{\text{scatt}}) \cdot \mathbf{r}_l(t_l)] \\ &\times S^*(\mathbf{k}_{\text{scatt}} - \mathbf{k}_{jl}, \tau) S^*(\mathbf{k}_{jl} - \mathbf{k}_{\text{inc}}, \tau). \end{aligned} \quad (23)$$

5. Electric-Field Correlation Functions

In this section we consider two closely spaced detectors, α and β , in the scattering far zone, as in Fig. 2. The angle between the detectors is δ , and their fields of view within the scattering cell almost completely overlap, i.e.,

$$D_\alpha(\mathbf{r}) \approx D_\beta(\mathbf{r}) \equiv D(\mathbf{r}). \quad (24)$$

We consider the two electric-field cross-correlation functions,

$$[c(\tau)] = \int_{-\infty}^{\infty} dT [E_{\alpha}^*(T + \tau) E_{\beta}(T)], \quad (25)$$

$$[c'(\tau)] = \int_{-\infty}^{\infty} dT [E_{\alpha}(T + \tau) E_{\beta}(T)], \quad (26)$$

where the electric field at each detector for each of the four input-output polarization channels is the sum of single- and double-scattering contributions. The correlation function $[c(\tau)]$ is dominant for autocorrelation¹ (i.e., $\delta = 0$), and $[c'(\tau)]$ is dominant for cross correlation for which two counterpropagating beams and two oppositely positioned detectors are used.¹⁷

The T integral in Eqs. (25) and (26) is modeled by the ensemble average of the integrands $\langle [E_{\alpha}^*(\tau) E_{\beta}(0)] \rangle$ and $\langle [E_{\alpha}(\tau) E_{\beta}(0)] \rangle$, respectively, i.e., the product of the fields is averaged over all the possible positions of particles j and l at times t_j and t_l and times $t_j + \tau$ and $t_l + \tau$ when scattering occurs.¹⁴ Averaging over the particle positions at $t_j + \tau$ and $t_l + \tau$, given the positions at t_j and t_l , was performed in Sections 3 and 4 and led to the scattering structure factors. We now average over the positions of the particles at t_j and t_l . For the single-scattering contribution to the electric-field correlation function $[c(\tau)]$, substitution of Eqs. (16) and (18) into Eq. (25) gives³³

$$\begin{aligned} [c_1(\tau)] &\approx \langle [E_{\alpha 1}^*(\tau) E_{\beta 1}(0)] \rangle \\ &= \frac{E_0^2 A_1^2 [P_1^2]}{k^2 R_d^2} \exp(i\omega\tau) S(\mathbf{\kappa}^{\alpha}, \tau) \\ &\times \left\{ \rho \int_{V_0} d^3 r_j |B(\mathbf{r}_j) D(\mathbf{r}_j)|^2 \right. \\ &\times \exp[-(L_{bj} + L_{jd})/L_{\text{scatt}}] \exp(i\mathbf{\epsilon} \cdot \mathbf{r}_j) \\ &+ \rho^2 \int_{V_0} d^3 r_j \int_{V_0} d^3 r_{jm} B^*(\mathbf{r}_j) D^*(\mathbf{r}_j) B(\mathbf{r}_m) D(\mathbf{r}_m) \\ &\times [g(r_{jm}) - 1] \exp[-(L_{bj} + L_{jd} + L_{bm} + L_{md})/2L_{\text{scatt}}] \\ &\times \exp(i\mathbf{\epsilon} \cdot \mathbf{r}_j) \exp(-i\mathbf{\kappa}^{\beta} \cdot \mathbf{r}_{jm}) \\ &+ \rho^2 \int_{V_0} d^3 r_j B(\mathbf{r}_j) D(\mathbf{r}_j) \exp[-(L_{bj} + L_{jd})/2L_{\text{scatt}}] \\ &\times \left. \exp(-i\mathbf{\kappa}_{\text{ave}} \cdot \mathbf{r}_{jm}) \right\}^2, \quad (27) \end{aligned}$$

where the subscript 1 denotes single scattering, the elements of $[P_1^2]$ are the squares of the elements of $[P_1]$, and

$$\begin{aligned} \mathbf{\epsilon} &= \mathbf{k}_{\text{scatt}}^{\alpha} - \mathbf{k}_{\text{scatt}}^{\beta} = \mathbf{\kappa}^{\alpha} - \mathbf{\kappa}^{\beta}, \\ |\mathbf{\epsilon}| &= 2k \sin(\delta/2), \\ \mathbf{\kappa}_{\text{ave}} &= (\mathbf{\kappa}^{\alpha} + \mathbf{\kappa}^{\beta})/2, \quad (28) \end{aligned}$$

where $\mathbf{k}_{\text{scatt}}^{\alpha, \beta}$ is the scattered wave vector from the origin to detector α or β and $\mathbf{\kappa}^{\alpha, \beta}$ is the scattered momentum transfer to α or β . The first term of approximation (27) is the single-particle contribution. The second term is the particle-pair contribution where $g(r_{jm})$ is the static pair-correlation function. The third term represents diffraction from the collection of particles taken as a whole. As we are not concerned with forward scattering, we ignore the diffraction term. Similarly, we limit our treatment to volume fractions $\phi \leq 0.1$ and do not consider critical opalescence near phase transitions.⁷ In this case, the static pair-correlation function is³⁴ $g(r_{jm}) \approx 1$ for $r_{jm} \geq 2a$, and we ignore pair-correlation effects. The remaining single-particle contribution to $[c_1(\tau)]$ may be written as

$$[c_1(\tau)] \approx \frac{E_0^2 A_1^2 N_0}{k^2 R_d^2} \exp(i\omega\tau) U_{\text{auto}} G_0(\mathbf{\epsilon}) [P_1^2] S(\mathbf{\kappa}^{\alpha}, \tau). \quad (29)$$

The autocorrelation strength factor,

$$U_{\text{auto}} \equiv \frac{1}{V_0} \int_{V_0} d^3 r_j |B(\mathbf{r}_j) D(\mathbf{r}_j)|^2 \exp[-(L_{bj} + L_{jd})/L_{\text{scatt}}], \quad (30)$$

contains both the roll-off of the transverse beam profile and variations in detector sensitivity in V_0 . The dimensionless geometric factor,

$$\begin{aligned} G_0(\mathbf{\epsilon}) &\equiv \frac{1}{U_{\text{auto}} V_0} \int_{V_0} d^3 r_j |B(\mathbf{r}_j) D(\mathbf{r}_j)|^2 \\ &\times \exp[-(L_{bj} + L_{jd})/L_{\text{scatt}}] \exp(i\mathbf{\epsilon} \cdot \mathbf{r}_j), \quad (31) \end{aligned}$$

is normalized to unity at $\mathbf{\epsilon} = 0$, which corresponds to autocorrelation.

To appreciate the significance of the geometric factor of Eq. (31), we consider V_0 to be a cylinder of radius R_y and length $2R_z$ oriented along the z axis, as in Fig. 2, which crudely models a focused laser beam in the vicinity of its focal waist. The length $2R_z$ is the smaller of either the diameter of the scattering cell or the diameter of the detector field of view. For simplicity in our analysis, we take $B(\mathbf{r})$ to be a hard-edge box function. The detector field of view is also assumed to be uniform, although our results remain valid for all realistic beam and detector sensitivity profiles (e.g., Gaussians²⁰). If the suspended particles are small, if they are approximately index matched to the liquid, or if their volume fraction is small, we can ignore the beam attenuation factors and obtain $U_{\text{auto}} = 1$. Under these assumptions, Eq. (31) becomes the Fourier transform of V_0 , i.e.,

$$G_0(\mathbf{\epsilon}) = \left[\frac{\sin(\epsilon R_z \cos \xi)}{\epsilon R_z \cos \xi} \right] \left[\frac{2J_1(\epsilon R_y \sin \xi)}{\epsilon R_y \sin \xi} \right], \quad (32)$$

where $\mathbf{\epsilon}$ in general lies in the yz plane and makes an angle ξ with the z axis. Our neglect of the beam attenuation factors is justified for only rather dilute suspensions because $R_z = 5$ mm for the experiment of Ref. 27 and $L_{\text{scatt}} \geq 3$ mm for $\phi \leq 10^{-3}$.

Equation (32) may be rewritten as

$$G_0(\epsilon) = \left[\frac{\sin(\delta \cos \xi / \delta_{\text{coh}}^z)}{\delta \cos \xi / \delta_{\text{coh}}^z} \right] \left[\frac{2J_1(\delta \sin \xi / \delta_{\text{coh}}^y)}{\delta \sin \xi / \delta_{\text{coh}}^y} \right], \quad (33)$$

where δ_{coh}^y and δ_{coh}^z have the following meanings. If the single-scattering volume is replaced by an incoherent light source with the same dimensions, the spatial coherence area of the source far down the x axis has the angular diameter³⁵

$$\delta_{\text{coh}}^y = \frac{1}{kR_y} \quad (34)$$

in the y direction and

$$\delta_{\text{coh}}^z = \frac{1}{kR_z} \quad (35)$$

in the z direction. Thus light singly scattered from within V_0 is strongly cross correlated {i.e., $G_0(\epsilon)$ and $[c_1(\tau)]$ are large} if detectors α and β lie within the coherence area of the analogous light source. In Section 6 we find a similar geometric factor with a similar interpretation in the double-scattering contribution to the cross-correlation function.

To this point we have tacitly assumed that detectors α and β are immersed in the liquid along with the suspended particles so that the ray paths from the scattering sites to the detectors are straight lines. Because the detectors in Fig. 2 lie outside the index-matching vat, the ray paths refract at the vat-air interface, thus changing the dimensions of the spatial coherence area at the detector plane. Because the scattered rays in the xy plane encounter a flat vat-air interface, a ray-tracing calculation gives

$$\delta_{\text{coh}}^y = \frac{\lambda}{2\pi R_y} \left[1 - \frac{(n_L - 1)R_v}{n_L R_d} \right], \quad (36)$$

where R_v is the radius of the vat. The last factor in Eq. (36) is due to refraction, while the fact that only a portion of the ray path is in the liquid removes the implicit n_L factor from k in the denominator of Eq. (34). Because the scattered rays in the xz plane encounter a curved vat-air interface, a similar ray-tracing calculation gives

$$\delta_{\text{coh}}^z = \frac{\lambda}{2\pi n_L R_z}, \quad (37)$$

which is identical to Eq. (35). Refraction at the curved interface introduces a new $1/n_L$ factor, while the fact that only a portion of the ray path is in the liquid removes the implicit $1/n_L$ factor from k in Eq. (35).

By a derivation similar to that which led to approximation (29), the single-scattering contribution to the second electric-field cross-correlation function $[c'(\tau)]$

of Eq. (26) is

$$\begin{aligned} [c_1'(\tau)] &\approx \langle [E_{\alpha 1}(\tau) E_{\beta 1}(0)] \rangle \\ &\approx \frac{E_0^2 A_1^2 N_0}{k^2 R_d^2} \exp(2ikR_d - i\omega\tau) [P_1^2] \\ &\quad \times S^*(\mathbf{k}_{\text{ave}}, \tau) U_{\text{auto}} G_0^*(2\mathbf{k}_{\text{ave}}). \end{aligned} \quad (38)$$

The geometric factor $G_0^*(2\mathbf{k}_{\text{ave}})$ greatly suppresses $[c_1'(\tau)]$ with respect to $[c_1(\tau)]$ for the one-beam cross-correlation geometry. For the cylindrical scattering volume of Fig. 2 and $\theta = 90^\circ$, $\lambda = 0.5145 \mu\text{m}$, and $R_y = 44 \mu\text{m}$, which correspond to the experimental parameters of Ref. 27, we obtain $G_0^*(2\mathbf{k}_{\text{ave}}) \approx 5 \times 10^{-5}$. If, on the other hand, detectors α and β are placed sufficiently close together, we have $G_0(\epsilon) \approx 1$ in $[c_1(\tau)]$. A similar level of suppression occurs for double scattering as well, and we no longer consider the electric-field cross-correlation function $[c'(\tau)]$ for the one-beam cross-correlation geometry.

For the double-scattering ensemble average, again we both ignore diffraction in the forward direction and assume that the particles have a sufficiently low volume fraction so that static pair-correlation effects are unimportant. We then obtain

$$\begin{aligned} [c_2(\tau)] &\approx \frac{E_0^2 A_2^2 \rho^2}{k^2 R_d^2} \exp(i\omega\tau) \int_{V_1} d^3 r_j |B(\mathbf{r}_j)|^2 \\ &\quad \times \exp(i\epsilon \cdot \mathbf{r}_j) \int_{V_2} d^3 r_{jl} \frac{|D(\mathbf{r}_l)|^2}{k^2 r_{jl}^2} [P_2^2] \\ &\quad \times S(\mathbf{k}_{\text{scatt}}^\alpha - \mathbf{k}_{jl}, \tau) S(\mathbf{k}_{jl} - \mathbf{k}_{\text{inc}}, \tau) \\ &\quad \times \exp[-(L_{bj} + r_{jl} + L_{ld})/L_{\text{scatt}}] \exp(i\epsilon \cdot \mathbf{r}_{jl}). \end{aligned} \quad (39)$$

Similarly, for the single-double-scattering cross terms we obtain

$$\begin{aligned} [c_{12}(\tau)] &\approx \langle [E_{\alpha 1}^*(\tau) E_{\beta 2}(0)] \rangle \\ &\approx \frac{E_0^2 A_1 A_2 \rho^2}{k^2 R_d^2} \exp(i\omega\tau) S(\mathbf{k}^\alpha, \tau) \int_{V_0} d^3 r_j |B(\mathbf{r}_j)|^2 \\ &\quad \times D^*(\mathbf{r}_j) \exp(i\epsilon \cdot \mathbf{r}_j) \int_{V_2} d^3 r_{jl} \frac{D(\mathbf{r}_l)}{kr_{jl}} [P_1 P_2] \\ &\quad \times \exp[-(2L_{bj} + L_{jd} + r_{jl} + L_{ld})/2L_{\text{scatt}}] \\ &\quad \times \exp[i(\mathbf{k}_{jl} - \mathbf{k}_{\text{scatt}}^\beta) \cdot \mathbf{r}_{jl}], \end{aligned} \quad (40)$$

$$\begin{aligned} [c_{21}(\tau)] &\approx \langle [E_{\alpha 2}^*(\tau) E_{\beta 1}(0)] \rangle \\ &\approx \frac{E_0^2 A_1 A_2 \rho^2}{k^2 R_d^2} \exp(i\omega\tau) \int_{V_0} d^3 r_j |B(\mathbf{r}_j)|^2 \\ &\quad \times D^*(\mathbf{r}_j) \exp(i\epsilon \cdot \mathbf{r}_j) \int_{V_2} d^3 r_{jl} \frac{D(\mathbf{r}_l)}{kr_{jl}} [P_1 P_2] \\ &\quad \times S(\mathbf{k}_{\text{scatt}}^\alpha - \mathbf{k}_{jl}, \tau) S(\mathbf{k}_{jl} - \mathbf{k}_{\text{inc}}, \tau) \\ &\quad \times \exp[-(2L_{bj} + L_{jd} + r_{jl} + L_{ld})/2L_{\text{scatt}}] \\ &\quad \times \exp[-i(\mathbf{k}_{jl} - \mathbf{k}_{\text{scatt}}^\alpha) \cdot \mathbf{r}_{jl}]. \end{aligned} \quad (41)$$

In approximations (40) and (41), the first of the two scatterings in $E_{\beta 2}$ or $E_{\alpha 2}^*$ occurs in volume V_0 rather than V_1 because the particle involved also contributes to the single-scattered electric field $E_{\alpha 1}^*$ or $E_{\beta 1}$, respectively. The single-triple-scattering cross-term contributions to the electric-field correlation function are of the same order³⁶ in A_1 as is the double-scattering contribution of approximation (39). These cross terms are not considered here.

6. Intensity Correlation Function

In this section we consider the intensity cross-correlation function

$$[C(\tau)] = \int_{-\infty}^{\infty} dT [I_{\alpha}(T + \tau) I_{\beta}(T)], \quad (42)$$

which is constructed from experimental measurements and is theoretically modeled by the ensemble average,

$$[C(\tau)] \approx \langle [E_{\alpha}^*(\tau) E_{\alpha}(\tau) E_{\beta}^*(0) E_{\beta}(0)] \rangle, \quad (43)$$

where each electric field contains both single-scattering and double-scattering contributions. By using Wick's theorem to expand approximation (43) in products of electric-field cross-correlation functions,¹⁷ we obtain

$$[C(\tau)] = [I_{\alpha}^{\text{ave}} I_{\beta}^{\text{ave}}] + [Y^* Y] + [W^* W], \quad (44)$$

where

$$[Y] = \langle [E_{\alpha 1}(\tau) E_{\beta 1}(0)] \rangle + \langle [E_{\alpha 1}(\tau) E_{\beta 2}(0)] \rangle + \langle [E_{\alpha 2}(\tau) E_{\beta 1}(0)] \rangle + \langle [E_{\alpha 2}(\tau) E_{\beta 2}(0)] \rangle, \quad (45)$$

$$[W] = \langle [E_{\alpha 1}^*(\tau) E_{\beta 1}(0)] \rangle + \langle [E_{\alpha 1}^*(\tau) E_{\beta 2}(0)] \rangle + \langle [E_{\alpha 2}^*(\tau) E_{\beta 1}(0)] \rangle + \langle [E_{\alpha 2}^*(\tau) E_{\beta 2}(0)] \rangle, \quad (46)$$

The $[I_{\alpha}^{\text{ave}} I_{\beta}^{\text{ave}}]$ term in Eq. (44) is the baseline of the intensity cross-correlation function, and the time dependence is contained in the $[Y^* Y]$ and $[W^* W]$ terms. As was seen in Section 5, we neglect the $[Y^* Y]$ term with respect to $[W^* W]$. Because both single and multiple scattering contribute to I_{α}^{ave} and I_{β}^{ave} whereas only single scattering is found in this section to contribute to $[W^* W]$, the signal-to-baseline value for cross correlation is expected to be smaller than that for autocorrelation, in which both single and multiple scattering contribute, albeit with different time dependences, to $[W^* W]$.

For previous autocorrelation and cross-correlation geometries that used collimated detectors, some of the integrations over particle coordinates in the double-scattering contribution to the autocorrelation function could not be performed analytically.⁵ We now assume that each particle j within the beam and the detector field of view (except those near the walls of the scattering cell) is surrounded by an isotropic environment of other particles l , also in the detector field of view, out to some average radius $r_{jl} \approx R_{\text{ave}}$, which is comparable with R_z . As was mentioned in Ref. 37, the isotropic

environment assumption is appropriate for a narrow beam and wide-field-of-view detectors. In this section it is shown that this assumption, although only approximately true for the geometry of Fig. 2, allows us to calculate analytically the double-scattering contribution to the autocorrelation and cross-correlation functions.

The isotropic environment assumption, the modeling of $B(\mathbf{r})$ and $D(\mathbf{r})$ by hard-edge box functions, and the neglect of the attenuation factors approximately decouple the $d^3 r_j$ and $d^3 r_{jl}$ integrations for the one-beam cross-correlation geometry. The $d^3 r_j$ integral in approximations (39)–(41) is identical to the single-scattering case. The double-scattering electric-field cross-correlation function of approximation (39) for noninteracting particles can then be written as

$$[c_2(\tau)] = \frac{E_0^2 A_1^2 N_0}{k^2 R_d^2} \exp(i\omega\tau) Q_2 U_{\text{auto}} G_0(\epsilon) [S_2(\theta, \tau, \epsilon)], \quad (47)$$

where

$$Q_2 = 3\phi \left(\frac{n^2 - 1}{n^2 + 2} \right)^2 (ka)^3 (kR_{\text{ave}}), \quad (48)$$

and the remaining integrations over particle coordinates are contained in

$$\begin{aligned} [S_2(\theta, \tau, \epsilon)] &= \begin{bmatrix} S_2^{VV} & S_2^{VH} \\ S_2^{HV} & S_2^{HH} \end{bmatrix} \\ &= \frac{1}{4\pi R_{\text{ave}}} \int_0^{R_{\text{ave}}} dr_{jl} \int_0^{\pi} \sin \theta_{jl} d\theta_{jl} \int_0^{2\pi} d\phi_{jl} [P_2^2] \\ &\quad \times \exp(-|\mathbf{k}_{\text{scatt}} - \mathbf{k}_{jl}|^2 D\tau) \\ &\quad \times \exp(-|\mathbf{k}_{jl} - \mathbf{k}_{\text{inc}}|^2 D\tau) \exp(i\epsilon \cdot \mathbf{r}_{jl}), \end{aligned} \quad (49)$$

where D is the diffusion coefficient of the particles in the liquid-filled scattering cell. Equation (47) differs from the single-scattering electric-field cross-correlation function of approximation (29) by the replacement of $[P_1^2]S$ by $Q_2[S_2]$.

The integrations in Eq. (49) may be performed analytically in the $\epsilon = 0$, $\epsilon R_{\text{ave}} \gg 1$, and $\tau = 0$ limits by use of a coordinate system rotated from the coordinate system of Fig. 4 by an angle $\gamma \equiv \theta/2$ about the y axis. The result for $\epsilon = 0$ (i.e., autocorrelation) is

$$S_2^{VV}(\tau) = \exp(-4k^2 D\tau) \left[\frac{3}{8} Z_0(b) + \frac{2}{8} Z_1(b) + \frac{3}{8} Z_2(b) \right], \quad (50)$$

$$\begin{aligned} S_2^{VH}(\tau) &= S_2^{HV}(\tau) = \exp(-4k^2 D\tau) [\cos^2 \gamma Z_0(b) \\ &\quad - (6 \cos^2 \gamma - 4) Z_1(b) \\ &\quad + (5 \cos^2 \gamma - 4) Z_2(b)], \end{aligned} \quad (51)$$

$$S_2^{HH}(\tau) = \exp(-4k^2 D\tau) \left[\left(\frac{19}{8} \cos^4 \gamma - 3 \cos^2 \gamma + 1 \right) \times Z_0(b) - \left(\frac{14}{8} \cos^4 \gamma - 4 \cos^2 \gamma + 2 \right) \times Z_1(b) + \left(\frac{3}{8} \cos^4 \gamma - \cos^2 \gamma + 1 \right) Z_2(b) \right] \quad (52)$$

where

$$b = 4k^2 D\tau \cos \gamma, \quad (53)$$

$$Z_m(b) \equiv \frac{1}{2} \int_{-1}^1 u^{2m} du \exp(bu) \quad (54)$$

for $m = 0, 1, 2$. The $Z_m(b)$ are combinations of hyperbolic sines and cosines divided by powers of b . These autocorrelation results merit the following comments. Equation (49) with $\epsilon = 0$ was first considered in Ref. 2 in a coordinate system in which the incident beam propagated in the $-x$ direction, the detector was located in the xy plane, and the integrations were performed numerically. The slope of Eqs. (50)–(52) at $\tau = 0$ is seen here to be $-4k^2 D$ and is independent of θ , a result first obtained in Ref. 2. Similarly, the graph of Eq. (51) is identical to the graph of $S_2^{VH}(\tau)$ shown in Fig. 2 of Ref. 2, which was obtained by means of numerical integration. The equality of $S_2^{VH}(\tau)$ and $S_2^{HV}(\tau)$ obtained here was first inferred on the basis of numerical integration in Ref. 3. The results of Eqs. (47)–(54) assume a point detector. For a finite-size detector, our results must be integrated with respect to ϵ over the detector face.²³ For a 3- μm -diameter monomode optical fiber coupled to a detector, ϵR_{ave} remains less than ~ 1.0 over the fiber cross section and the results of Eqs. (47)–(54) remain valid.

When ϵ is in the y direction, as in Fig. 2, and $\epsilon R_{\text{ave}} \gg 1$ corresponds to the cross correlation of two closely spaced detectors (e.g., a 250- μm detector spacing at $x = 16$ cm, as in Ref. 27, gives $\delta \approx 10^{-3}$ rad or $\epsilon R_{\text{ave}} \approx 100$), we have

$$S_2^{VV} = \frac{\pi}{2\epsilon R_{\text{ave}}} \exp(-4k^2 D\tau) I_0(b), \quad (55)$$

$$S_2^{VH} = S_2^{HV} = O[(\epsilon R_{\text{ave}})^{-3}], \quad (56)$$

$$S_2^{HH} = \frac{\pi}{2\epsilon R_{\text{ave}}} \exp(-4k^2 D\tau) \times \left[\cos^4 \gamma I_0(b) - \frac{2 \cos^2 \gamma}{b} I_1(b) + \frac{3}{b^2} I_2(b) \right], \quad (57)$$

where $I_m(b)$ for $m = 0, 1, 2$ are modified Bessel functions. These cross-correlation results merit the following observations. Whereas the single-scattering correlation function of approximation (29) is exponential in τ for noninteracting particles, the time dependence of $Z_m(b)$ for autocorrelation and $I_m(b)$ for cross

correlation causes the double-scattering contribution to the electric-field correlation functions to be nonexponential in τ for $\theta \neq 180^\circ$. Also, although there is substantial double scattering in the VH and the HV polarization channels for autocorrelation, double-scattering cross correlation is exceedingly weak in these channels.

Again when ϵ is in the y direction, calculation of the ϵ dependence of $[S_2]$ appears to be relatively simple in only the $\tau = 0$ limit (i.e., the intensity ensemble average). In this limit we obtain

$$S_2^{VV} = \frac{8}{\epsilon R_{\text{ave}}} \int_0^{\epsilon R_{\text{ave}}} du \frac{j_2(u)}{u^2}, \quad (58)$$

$$S_2^{VH} = S_2^{HV} = \frac{1}{\epsilon R_{\text{ave}}} \int_0^{\epsilon R_{\text{ave}}} du \left[\frac{j_1(u)}{u} - \frac{4j_2(u)}{u^2} \right], \quad (59)$$

$$S_2^{HH} = \frac{1}{\epsilon R_{\text{ave}}} \int_0^{\epsilon R_{\text{ave}}} du \left\{ \cos^2 \theta \left[j_0(u) - \frac{2j_1(u)}{u} \right] + (2 \cos^2 \theta + 1) \frac{j_2(u)}{u} \right\}, \quad (60)$$

where $j_m(u)$ for $m = 0, 1, 2$ are spherical Bessel functions. The $\epsilon = 0$ and $\epsilon R_{\text{ave}} \gg 1$ limits of Eqs. (58)–(60) agree with Eqs. (50)–(52) and Eqs. (55)–(57), respectively.

The single-double-scattering cross terms of approximations (40) and (41) may be written as

$$[c_{12}(\tau)] = \frac{E_0^2 A_1^2 N_0}{k^2 R_d^2} \exp(i\omega\tau) Q_{12} U_{\text{auto}} G_0(\epsilon) [S_{12}(\theta, \tau)], \quad (61)$$

where

$$Q_{12} = \frac{1}{2} \left(\frac{n^2 + 2}{n^2 - 1} \right) (ka)^{-6} (kR_{\text{ave}})^{-1} Q_2, \quad (62)$$

and the remaining integrations over particle coordinates are contained in

$$[S_{12}(\theta, \tau)] = \frac{k^2}{2\pi} S(\kappa^\alpha, \tau) \int_0^{R_{\text{ave}}} r_{jl} \exp(ikr_{jl}) dr_{jl} \times \int_0^\pi \sin \theta_{jl} d\theta_{jl} \int_0^{2\pi} d\phi_{jl} [P_1 P_2] \times \exp(-i\mathbf{k}_{\text{scatt}}^\beta \cdot \mathbf{r}_{jl}). \quad (63)$$

Similarly,

$$[c_{21}(\tau)] = \frac{E_0^2 A_1^2 N_0}{k^2 R_d^2} \exp(i\omega\tau) Q_{12} U_{\text{auto}} G_0(\epsilon) [S_{21}(\theta, \tau)], \quad (64)$$

where

$$\begin{aligned}
[S_{21}(\theta, \tau)] = & \frac{k^2}{2\pi} \int_0^{R_{\text{ave}}} r_{jl} \exp(-ikr_{jl}) dr_{jl} \int_0^\pi \sin \theta_{jl} d\theta_{jl} \\
& \times \int_0^{2\pi} d\phi_{jl} [P_1 P_2] \exp(-|\mathbf{k}_{\text{scatt}} - \mathbf{k}_{jl}|^2 D\tau) \\
& \times \exp(-|\mathbf{k}_{jl} - \mathbf{k}_{\text{inc}}|^2 D\tau) \exp(i\mathbf{k}_{\text{scatt}}^\alpha \cdot \mathbf{r}_{jl}).
\end{aligned} \quad (65)$$

The integration of Eqs. (63) and (65) shows that $[S_{12}]$ and $[S_{21}]$ are each large for $\epsilon R_{\text{ave}} \gg 1$. But they are opposite in sign and nearly equal in magnitude, and they almost completely cancel when added together in Eq. (46). The cancellation is maximal at $\tau = 0$ and slowly decreases for $\tau > 0$ because of the different combinations of structure factors in Eqs. (63) and (65). The fact that $Q_{12} \approx 10^{-4} Q_2$ for $n = 1.2$, $ka = 0.87$, and $R_{\text{ave}} \approx 4$ mm, as in Ref. 27, further decreases the importance of the cross terms for both autocorrelation and cross correlation. The cross terms have also been found to be unimportant for a number of other scattering geometries.^{3,17,36,38,39}

The suppression of double scattering and, by inference, the suppression of all multiple scattering for a single focused laser beam propagating through the scattering cell and cross correlation of two closely spaced detectors, as in Refs. 27–29, can be demonstrated as follows. For $a = 0.053$ μm polystyrene latex spheres in water²⁷ with $n = 1.2$, $\lambda = 0.5145$ μm , and $R_{\text{ave}} \approx 4$ mm, the product of the factors $Q_2[S_2]$ in Eq. (47) becomes larger than unity at $\tau = 0$ for autocorrelation, i.e., the double-scattering contribution becomes larger than the single-scattering contribution for $\phi \geq 10^{-3}$. This is experimentally observed in Ref. 27. The time dependence of the autocorrelation function for noninteracting particles ceases being exponential at volume fractions of this order both because $[S_2]$ is nonexponential in τ and, perhaps more importantly, because the general rate of falloff of $[S_2]$ differs from that of S . On the other hand, the profile of the single-scattering spatial coherence area as a function of the detector angular separation δ in the four polarization channels is modeled by $G_0(\epsilon) [P_1^2] S$ in Eqs. (28) and approximation (29). Similarly the profile of the double-scattering spatial coherence area as a function of δ in the four polarization channels is modeled by $G_0(\epsilon) [S_2(\theta, \tau, \epsilon)]$ in Eq. (47). The amount of double scattering for cross correlation with $\epsilon R_{\text{ave}} \gg 1$ relative to the amount of double scattering for autocorrelation with $\epsilon = 0$ in the VV polarization channel at $\tau = 0$ is

$$F^{VV}(\delta) = \frac{S_2^{VV}(\theta, \tau = 0, \epsilon R_{\text{ave}} \gg 1)}{S_2^{VV}(\theta, \tau = 0, \epsilon = 0)} = \frac{4\pi}{15\epsilon R_{\text{ave}}}, \quad (66)$$

If detectors α and β are placed so that $\delta \approx 1/kR_y$, singly scattered light is strongly correlated and $F^{VV}(\delta)$ becomes

$$F^{VV}(\delta) \approx \frac{4\pi R_y}{15R_{\text{ave}}} \approx 0.009 \quad (67)$$

for the values of R_y and R_{ave} in Ref. 27, thus reducing the double-scattering amplitude 100-fold from its autocorrelation value. This degree of suppression is comparable with the value calculated by Schätzel for the two-color cross-correlation system.²⁰ The τ dependence of $F^{VV}(\delta)$ is obtained by the division of Eq. (55) by Eq. (50) but does not significantly affect the degree of suppression from the value given in approximation (67). As a result, the intensity cross-correlation function of Eqs. (44) and (46) for the one-beam cross-correlation geometry is dominated by single scattering and remains exponential in τ for noninteracting particles at values of ϕ for which the autocorrelation function is no longer exponential.

Experimentally it is seen in Ref. 27 that multiple-scattering suppression occurs for the geometry of Fig. 2 for $\phi \leq 0.1$, which is far beyond the range of validity of the assumptions leading to approximation (67). At high volume fractions, triple scattering and beyond become important. Particle l is not necessarily in the far zone of particle j so that the Lorenz–Mie near-zone scattering amplitude must be employed. Similarly, pair-correlation effects, particle interaction effects, and the beam attenuation factors cannot be ignored. Nonetheless, the experimentally observed multiple-scattering suppression at large volume fractions attests to the robustness of the one-beam cross-correlation technique.

7. Conclusions

The principal result is that for the one-beam cross-correlation system, multiple scattering at low volume fractions occurs over a relatively large region in the y direction, whereas single scattering occurs within only the focused laser beam. As a result, multiple scattering produces a smaller far-zone coherence length in the y direction than does single scattering. If detectors α and β lie on the same single-scattering coherence area but on different multiple-scattering coherence areas, single scattering should be strongly cross correlated but multiple scattering should not be. Although this result may be thought of as being intuitively obvious in retrospect, it is pleasing to see that it can be derived with standard light-scattering theory.

The development presented here also fills in various details that the intuitive spatial coherence area argument alone cannot provide, such as the polarization dependence of the autocorrelation and cross-correlation results and the degree of spatial coherence as a function of δ , which is of special interest in light of the experimental results of Ref. 29. Our analytical evaluation of the τ dependence of the double-scattering contribution to the autocorrelation function, although dependent on the isotropic envi-

ronment assumption that is only approximately satisfied, is also of interest. This analytical result, if extendable to higher orders of multiple scattering, should provide a method for analyzing the experimental autocorrelation results of Ref. 40 that span the particle volume fraction range from the semidilute limit to the deep multiple-scattering regime. Previous multiple-scattering analyses of autocorrelation spectra assumed that each multiparticle-scattering contribution was an exponential with a different decay rate.⁶ Nonexponential contributions, such as those derived here for double scattering, should more realistically describe the experimental spectra.

The real assessment of the importance of the one-beam cross-correlation technique is the ease of its alignment, the rate at which and the level to which noise in the cross correlogram is decreased, and the simplicity of the analysis of the cross correlogram. These issues are addressed separately.²⁷⁻²⁹

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