Comments

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on "Polarization memory of multiply scattered light"

Isaac Freund and Moshe Kaveh
Department of Physics, Bar-Ilan University, Ramat-Gan 52 900, Israel
(Received 22 December 1989)

Recent data for the time correlation function for multiply scattered light in reflection are shown to be universal.

In a recent paper,[1] data for the time correlation function for multiply scattered light in reflection[2–7] are presented as a function of particle size and optical polarization. In the penultimate sentence of their introduction, the authors of Ref. 1 claim their data "show that contrary to previous reports (their Ref. 9, our Ref. 3), the form of the autocorrelation functions is not universal, but instead depends on both particle size and polarization." Is this correct, or do these data actually *support* the universality that we reported previously?[3]

Our claim of universality was based upon a scalar-wave treatment of the optical field and its agreement with our experiments.[3] What is the application of this scalar theory (if any) to a vector optical field? For the time correlation problem, the major differences between scalar and vector fields is the different relative weights of the Feynman paths associated with transport of the multiply scattered light. Different polarization channels of the vector field weigh these paths differently. For a linearly polarized input, for example, the parallel, copolarized output channel weighs short, polarization preserving paths more heavily than does the scalar theory, while the perpendicular, crosspolarized channel places greater relative weight on the long, depolarizing paths. Clearly then, the closest equivalent to the scalar theory is to collect both channels simultaneously, i.e., to simply collect all the multiply scattered light *without any polarization bias.* This is what we did in our previous experiments.[3] We demonstrate here that when this approach is applied to the data of Ref. 1, also these data confirm the system independence of the unbiased time correlation function in reflection, which we reported previously[3] based on scalar theory.

Consider as an example a linearly polarized input field. Denoting the copolarized output channel by the subscript $\parallel$, and the crosspolarized channel by the subscript $\perp$, the unbiased (i.e., total) intensity $I(t)$ is simply

$$I(t)=I_{\parallel}(t)+I_{\perp}(t) .$$

Since the fluctuations in the two orthogonal output channels are uncorrelated at all times, we may write the unbiased (i.e., equal weight for both channels) electric-field time correlation function $\langle G_1(t)\rangle$ as

$$\langle G_1(t)\rangle=[\langle I_{\parallel}\rangle G_1(t,\gamma_{\parallel}) + \langle I_{\perp}\rangle G_1(t,\gamma_{\perp})]/[\langle I_{\parallel}\rangle+\langle I_{\perp}\rangle] , \quad (1)$$

where γ parametrizes the differences between the two channels. $\langle G_1\rangle$ may, for example, be measured in a heterodyne experiment using a polarizer at 45° to combine with equal weight the optical fields from both channels. Working within the framework of scalar wave theory, Pine *et al.*,[5] and Edrei and Kaveh[6] have suggested for short times the compact form

$$G_1(t,\gamma)=\exp(-\gamma\sqrt{6t/\tau}) , \quad (2)$$

where τ is the single scattering correlation time. From Eqs. (1) and (2) we obtain $\langle\gamma_1\rangle$ of the unbiased electric-field time correlation function

$$\langle\gamma_1\rangle=[\langle I_{\parallel}\rangle\gamma_{\parallel}+\langle I_{\perp}\rangle\gamma_{\perp}]/[\langle I_{\parallel}\rangle+\langle I_{\perp}\rangle] . \quad (3)$$

In Table I, we list the results of the above analysis as applied to all the data of Ref. 1 for which both γ and $\langle I\rangle$ are reported. As may be seen, the same universal value of $\langle\gamma_1\rangle=2.06\pm0.03$ emerges for widely different particle sizes and for both linearly and circularly polarized input fields.

The unbiased intensity-intensity time correlation function $\langle G_2(t)\rangle$, which is easily measured by collecting all the scattered light with no polarization bias, is also well approximated by a stretched exponential

$$\langle G_2(t)\rangle=\exp(-2\langle\gamma_2\rangle\sqrt{6t/\tau}) , \quad (4)$$

where

$$\langle\gamma_2\rangle=[\langle I_{\parallel}\rangle^2\gamma_{\parallel}+\langle I_{\perp}\rangle^2\gamma_{\perp}]/[\langle I_{\parallel}\rangle^2+\langle I_{\perp}\rangle^2] . \quad (5)$$

Values for $\langle\gamma_2\rangle$ are given in Table I, from which it may be seen that also here the same universal value of $\langle\gamma_2\rangle=2.0\pm0.1$ emerges for the different particle sizes for linearly and circularly polarized light, while the average 5% dispersion matches well the stated 5% error limits of Ref. 1.

In Figs. 1 and 3 of Ref. 1, the γ's are plotted for a wider range of particle sizes than is listed in Table I, but the corresponding $\langle I\rangle$ are not reported, so these additional

TABLE I. Analysis of time correlation data. Data from Ref. 1.

Polarization state	Particle diam. (μm)	γ_p[a]	γ_q[a]	$\langle I_p \rangle / \langle I_q \rangle$[a]	$\langle \gamma_1 \rangle$[b]	$\langle \gamma_2 \rangle$[b]
Linear	0.091	1.45	3.06	1.78	2.03	1.84
	0.605	1.96	2.18	1.05	2.07	2.06
Circular	0.091	2.68	1.59	0.69	2.04	1.94
	0.605	1.72	2.62	1.40	2.10	2.02
					ave. = 2.06 ±0.03	ave. = 2.0 ±0.1

[a]The subscripts p and q refer to either the $\parallel$ and $\perp$ channels for linear, or the + and − channels for circular polarization.
[b]$\langle \gamma_1 \rangle$ is defined in Eq. (3), $\langle \gamma_2 \rangle$ in Eq. (5).

data points cannot presently be included in the table. Nonetheless, it is clear from the trends displayed in these two figures that all of the data *will* closely follow the results of Table I. Accordingly, we conclude that the data of Ref. 1 fully *support* the system independence we reported previously[3] based on scalar theory, so that these data make an important contribution by significantly enlarging the domain over which this system-independent behavior is verified experimentally.

We now note that even the individual γ's of the various polarization channels also are given by a simple empirical formula. Defining a depolarization factor

$$\rho = \langle I_{\text{opposite}} \rangle / \langle I_{\text{same}} \rangle \ , \qquad (6)$$

where the subscript "same" refers to an output polarization channel which is the same as that of the input field, while the subscript "opposite" refers to the opposite channel (i.e., for linear polarization, same denotes $\parallel$ and opposite denotes $\perp$, while for circular polarization same denotes + and opposite denotes −), we find

$$\gamma_{\text{opposite}} = \left[\frac{1+\rho}{\rho+\rho^{\alpha}} \right] \gamma_0 \ , \qquad (7a)$$

and

$$\gamma_{\text{same}} = \rho^{\alpha} \gamma_{\text{opposite}} \ , \qquad (7b)$$

where $\alpha = \frac{4}{3}$ and γ_0 is the scalar value of γ. In Table II we compare the measured γ with the results of Eqs. (7) using as the value of γ_0 appropriate to the data of Ref. 1, $\gamma_0 = 2.06$. As may be noted from this table, Eqs. (7) provide a nearly perfect description of the data for widely different particle sizes for both linear and circular polarizations.[8]

Although the data do exhibit the expected system-independent behavior, γ_0 is somewhat larger than the expected scalar theory value

$$\gamma_0 = 1 + \Delta = 1.7104 \ldots ,$$

where Δ, which arises from the boundary conditions for the diffusion equation, is obtained from Milne theory.[9] This implies that the measured $G_2(t)$ may be too narrow. We suggest here that the source of this narrowing is the insidious problem of internal surface reflections,[10–12] which act to reinject a portion of the scattered light. This reinjection leads to an artificial elongation of the Feynman paths traversed by the photon, and thus to an artificial narrowing of the time correlation function. Relevant here is the total fraction r of the scattered light, which is returned to the sample by these internal reflections. We note that because multiple scattering scrambles the incident beam direction, r can greatly exceed the typical 4% reflectivity for normal incidence.

There are two important sources of internal surface reflection. The potentially largest source is total internal reflection at the external glass-air interface of the sample cell. Assuming more-or-less isotropic emission of the diffusely scattered light, and taking account of refraction at the inner water-glass interface, we estimate that a little over half the scattered light is returned to the sample by this mechanism. This is a potentially significant problem.

TABLE II. Comparison of theory and experiment for γ.

Polarization state	Particle diam. (μm)	ρ[a]	Meas. γ_p[b]	γ_p[b]	Calc./meas.	Meas. γ_q[b]	Calc. γ_q[b]	Calc./meas.
Linear	0.091	0.56	1.45	1.45	1.00	3.06	3.15	1.03
	0.605	0.95	1.96	1.99	1.02	2.18	2.13	0.98
Circular	0.091	1.45	2.68	2.68	1.00	1.59	1.63	1.03
	0.605	0.71	1.72	1.67	0.97	2.62	2.61	1.00
				ave.	= 1.00 ±0.02		ave.	= 1.01 ±0.02

[a]$\rho = \langle I_\perp \rangle / \langle I_\parallel \rangle$ for linear, and $\langle I_- \rangle / \langle I_+ \rangle$ for circular polarization.
[b]The subscripts p and q refer to either the $\parallel$ and $\perp$ channels for linear, or the + and − channels for circular polarization.

When recognized in advance, it can be avoided by proper experimental technique. In uncontrolled experiments, however, it will manifest itself to a greater or lesser extent depending upon the precise details of the sample and cell geometries, the collecting optics, etc. For example, in the typical glass cell with windows a few mm in thickness, one usually sees a distinct, luminous ring which surrounds the central bright spot produced by the incident laser beam. This ring arises from the total internal reflections described above. If the laser beam diameter is less than the window thickness (it is not clear that this is true for Ref. 1), the luminous ring is well separated from the main spot and can be masked out, thereby avoiding the worst part of the problem. Even if this is done, however, one is still left with the lesser problem of light reemitted by the ring being partially reflected back into the central spot, so that either very thick cell windows, or some index matching technique[10] is required to eliminate the reflection problem altogether. With a large beam diameter (1 cm was used in Ref. 1) and thin cell windows, a substantial fraction of the scattered light is necessarily returned to the sample, thereby leading to a significant value for r and thus to a significant narrowing of the measured correlation function. Since no mention at all is made of these important effects in either Ref. 1, or in earlier work by this group,[5] and only minimal experimental details are reported, we are unable to provide a first-principle estimate of r for their experimental arrangement.

The second source of internal surface reflections is essentially unavoidable. This is the Fresnel reflection at the internal water-glass interface. Assuming equal amplitude s- and p-polarized components in the multiply scattered light, we estimate for this effect $r=0.12$, which sets the lower limit for this quantity.

We have recently shown[12] that for reflection from a thick sample, as in the experiments of Ref. 1, the apparent (measured) value of the time correlation function G_2' in the presence of internal reflections is related to the true value G_2 of the correlation in the absence of reflections, independent of the form of G_2, by

$$G_2'(r)=(1-r)^2G_2/[1-r\sqrt{G_2}]^2 . \tag{8}$$

Using the compact form Eq. (4) for G_2, this leads to the relationship between an apparent value γ', and a true value γ_0,

$$\gamma'=\gamma_0/(1-r) . \tag{9}$$

From Eq. (9) and the data of Table I, we are able to estimate that for the experiments of Ref. 1, $r=0.17$, so that surface reflections appear to play a significant role here, resulting in a half-width for G_2' which is almost 30% too small. It appears, then, that the absolute accuracy of 5% claimed in Ref. 1 may be prematurely optimistic. We note that, since r is a property of a given experimental apparatus, the system independence of the time correlation function is preserved for that apparatus, albeit with an erroneous value of γ. It is thus clear that in all future careful work special attention must be paid to minimizing internal surface reflections, and to properly correcting the data for the unavoidable reflections that do remain.[12]

In summary, we have shown that by simply collecting the scattered light without polarization bias, the universality of the time correlation function in reflection, which we had reported previously[3] based upon scalar theory, is fully preserved. Although the detailed, complicated behavior of particular polarization channels may be of some significance in certain instances, the universality of the unbiased time correlation function will clearly always be of primary importance. We have also shown that the individual polarization channels themselves follow a simple law which is expressed in terms of the easily measured depolarization ρ.

---

[1]F. C. MacKintosh, J. X. Zhu, D. J. Pine, and D. A. Weitz, Phys. Rev. B **40**, 9342 (1989).

[2]G. Maret and P. E. Wolf, Z. Phys. B **65**, 409 (1987).

[3]M. Rosenbluh, M. Hoshen, I. Freund, and M. Kaveh, Phys. Rev. Lett. **58**, 2754 (1987).

[4]M. J. Stephen, Phys. Rev. B **37**, 1 (1988).

[5]D. J. Pine, D. A. Weitz, P. M. Chaiken, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).

[6]I. Edrei and M. Kaveh, J. Phys. C **21**, L971 (1988).

[7]D. N. Qu and J. C. Dainty, Opt. Lett. **13**, 1066 (1988).

[8]We also find that

$$\frac{\langle I_+\rangle}{\langle I_-\rangle}=\frac{3}{2}\left[\frac{\langle I_\perp\rangle}{\langle I_\parallel\rangle}\right]^{4/3},$$

so that *everything* may be simply expressed in terms of the depolarization factor for *linearly* polarized light.

[9]P. M. Morse and H. Feschbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953).

[10]I. Freund, M. Rosenbluh, and R. Berkovitz, Phys. Rev. B **39**, 12 403 (1989).

[11]A. Lagendijk, R. Vreeker, and P. DeVries, Phys. Lett. A **136**, 81 (1989).

[12]I. Freund and R. Berkovitz, Phys. Rev. B **41**, 496 (1990); *ibid.* **41**, 9540(E) (1990).