Random Walk of Polarized Light in Turbid Media

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We study the propagation of polarized light in turbid media as a random walk of vector photons. Both propagation and polarization directions of light are found to isotropize, following a power law of the number of scattering events. The characteristic length scale governing light isotropization and linear depolarization, the isotropization length l_p, is derived using the exact Mie scattering for spherical particles. A simple relation $l_p/l_t \simeq 2.8 - 2.5g$ is obtained for Rayleigh-Gans scatterers where l_t is the transport mean free path and g is the mean cosine of scattering angles.

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The propagation of polarized light in random media is a difficult problem to handle in the framework of multiple-scattering theory. The classical approach, which assumes that phases are uncorrelated on scales larger than the scattering mean free path l_s, leads to the radiative transport equation in which any interference effects are neglected [1]. The remnant coherent effect of multiply scattered light survives in the backscattering cone [2,3]. Although a scalar-wave model for the optical field has been used successfully in many instances, it is becoming increasingly apparent that the vector nature of light plays an important role in diverse phenomena such as coherent backscattering [4,5], diffuse wave spectroscopy (DWS) [6–9], and memory effect [7,10]. Experiments and numerical simulations reveal strikingly different behavior of light depolarization by Rayleigh and non-Rayleigh scatterers [7,10–14]. Previous theoretical attempts have mainly focused on either Rayleigh scattering [4,15,16] or circular depolarization by large particles [17] in which simplifications were made about the softness of the particle and the scattering angles of light and restrict its application even for large scatterers.

Light scattering by particles neither very small nor large and soft contains a much more complex structure originated from Mie resonance scattering. Light depolarization by spherical particles with no restrictions in their size and refractive index, the so called Mie scatterers, is hence most challenging. The behavior of light depolarization by Mie scatterers has been found experimentally and numerically to be complex and sensitive to both the size and the refractive index of the particle [12–14]. A general rule of light depolarization by Mie scatterers is, however, hard to infer from these observations as "a different behavior of the depolarization of polarized light may arise due to the pronounced effects of resonance and the interference structures" [14] in Mie scattering. A rigorous theoretical analysis is required to answer the delicate dependence of light depolarization on the size and refractive index of Mie scatterers. Identifying the characteristic length scale over which light depolarization occurs is of ultimate importance since that length scale is a limiting factor in using polarized light in vast applications including remote sensing, biomedical imaging, and many other multidisciplinary fields where multiply scattered light is used to probe the interior of disordered media such as atmosphere, colloidal suspensions and biological tissues [6–8,18,19].

In this Letter, we analytically treat propagation of polarized light in a uniform turbid medium as a random walk of vector photons to give new insight into the underlying physics. Contrary to a random walk of scalar photons, each scattering of the photon is governed by the amplitude scattering matrix of the scatterer and generally alters both the propagation direction and the polarization. The free path between consecutive scattering events is assumed to be independent of the propagation direction and the polarization of light, following the tradition of radiative transfer. This independence enables us to study the microscopic statistics of photon propagation and depolarization in the direction space first and to analyze light depolarization in real space by mapping the number of scattering events later.

The key finding of this Letter is the existence and the analytical expression of the characteristic length scale, the isotropization length l_p, governing light isotropization and linear depolarization when propagating in a scattering medium. Light tends to lose its memory of the initial propagation direction during propagation over one transport mean free path l_t. We find light gradually becomes isotropic (both the propagation direction becomes isotropic and the intensity equalizes in all polarization directions) in the direction space during propagation in a scattering medium composed of spherical or randomly oriented nonspheric scatterers, characterized by the isotropization length l_p. The isotropization length is longer than l_t. The complex dependence of l_p on the size and refractive index of the scatterer for spherical particles reveals the pronounced effects of Mie resonance and interference structures. In addition, a simple relation $l_p/l_t \simeq 2.8 - 2.5g$ is obtained for small particles of modest relative refractive index with $g \lesssim 0.72$ where l_t is the transport mean free path and g is the mean cosine of scattering angles, extending the well-known result for Rayleigh scattering to the Rayleigh-Gans scattering regime.

We describe light scattering in a local orthonormal coordinate system $(\mathbf{p}, \mathbf{q}, \mathbf{s})$ where $\mathbf{p}$ and $\mathbf{q}$ are the unit vectors along the parallel and perpendicular electric field with respect to the scattering plane of the *previous* scattering and $\mathbf{s}$ is the photon's propagation direction prior to the current scattering. Light scattering by the particle can be described by a rotation of the local coordinate system $(\mathbf{p}', \mathbf{q}', \mathbf{s}')^T = A(\mathbf{p}, \mathbf{q}, \mathbf{s})^T$ and an update of the parallel and perpendicular electric field components $(E_1', E_2')^T = B(E_1, E_2)^T$ where the superscript "T" denotes transpose [20]. The matrices A and B are given by:

$$A = \begin{pmatrix} \mu\cos\phi & \mu\sin\phi & -\nu \\ -\sin\phi & \cos\phi & 0 \\ \nu\cos\phi & \nu\sin\phi & \mu \end{pmatrix} \tag{1}$$

and

$$B = \gamma^{-1/2}\begin{pmatrix} S_2\cos\phi & S_2\sin\phi \\ -S_1\sin\phi & S_1\cos\phi \end{pmatrix}, \tag{2}$$

where $\mu \equiv \cos\theta$ and $\nu \equiv \sin\theta$, θ and ϕ are the scattering and azimuthal angles, respectively, and $S_{2,1}$ are the nonzero elements of the amplitude scattering matrix of the spherical or randomly oriented nonspheric scatterers [21]. A scaling constant $\gamma \equiv \frac{1}{4}\int(|S_2|^2 + |S_1|^2)d\mu$ has been introduced such that the energy of light is conserved $(|E_1'|^2 + |E_2'|^2 = |E_1|^2 + |E_2|^2)$ in scattering on average. Any arbitrary scaling constant can be used here as it does not change either the polarization property of light or the phase function of scattering. Free flight of light in space can also be suppressed in light multiple scattering in the direction space as it does not change the polarization. The matrix A is orthogonal. The exact Mie scattering phase function is realized by weighting light scattering into different directions with the matrix B given a uniformly distributed scattering and azimuthal angles over the entire solid angle.

To analyze the second order statistics about light polarization and propagation directions in the direction space, we introduce $\mathbf{F} \equiv (E_1\mathbf{p}, E_1\mathbf{q}, E_1\mathbf{s}, E_2\mathbf{p}, E_2\mathbf{q}, E_2\mathbf{s})^T$ and consider

$$\mathbf{F}^{(n)} = C^{(n)}\mathbf{F}^{(n-1)} \tag{3}$$

in the nth scattering where the superscript "(n)" denotes the quantities after the nth scattering and $C^{(n)}$ is the direct product

$$C^{(n)} \equiv B \otimes A = \begin{pmatrix} B_{11}A & B_{12}A \\ B_{21}A & B_{22}A \end{pmatrix}, \tag{4}$$

with (θ, ϕ) the scattering and azimuthal angles in the nth scattering. After light is scattered n times, the electric field is given by $\mathbf{E}^{(n)} = \mathbf{F}_1^{(n)} + \mathbf{F}_5^{(n)}$ and the propagation direction is specified by $\mathbf{F}_3^{(n)}$ and $\mathbf{F}_6^{(n)}$.

Equation (3) can be regarded as the "equation of motion" of light in the direction space. To proceed, we use a transfer matrix technique [4,22] and write $\mathbf{F}^{(n)} = M^{(n)}\mathbf{F}^{(0)} = \prod_{i=1}^{n} C^{(i)}\mathbf{F}^{(0)}$ with

$$M^{(n)} = C^{(n)}M^{(n-1)}, \qquad M^{(0)} = I, \tag{5}$$

where I is the unit matrix. The motion of polarized light in the direction space is embodied in the transfer matrix $M^{(n)}$.

We start by considering an incident light of unit intensity polarized along the x axis and propagating in the positive z axis, i.e., $\mathbf{F}^{(0)} = (\hat{x}, \hat{y}, \hat{z}, 0, 0, 0)^T$. The electric field after n scattering events is given by $\mathbf{E}^{(n)} = (M_{11}^{(n)} + M_{51}^{(n)})\hat{x} + (M_{12}^{(n)} + M_{52}^{(n)})\hat{y} + (M_{13}^{(n)} + M_{53}^{(n)})\hat{z}$. Consider its x component $E_x^{(n)}$. Denote $\xi_j^{(n)} = M_{j1}^{(n)}(M_{j1}^{(n)})^*$ for $j = 1, 2, \ldots, 6$, $\xi_7^{(n)} = M_{11}^{(n)}(M_{51}^{(n)})^* + M_{51}^{(n)}(M_{11}^{(n)})^*$, and $\xi_8^{(n)} = M_{21}^{(n)}(M_{41}^{(n)})^* + M_{41}^{(n)}(M_{21}^{(n)})^*$. We find

$$\langle \boldsymbol{\xi}^{(n)} \rangle = G\boldsymbol{\xi}^{(n-1)}, \tag{6}$$

where G is an 8×8 transfer matrix and $\langle\rangle$ denotes the ensemble average on the scattering and azimuthal angles over the whole solid angle (the average of terms involving odd powers of $\sin\phi$ or $\cos\phi$ vanishes). Only the averaging over the scattering angles at the nth scattering event is performed in Eq. (6). Because of the specific structure of $C^{(n)}$ reflecting underlying symmetry, 4 out of 8 eigenvalues of G are zero. The nonzero eigenvalues are 1, d, and $\lambda_\pm = \frac{3}{4} + \frac{d}{4} - \frac{7}{16}(a + b) \pm \frac{1}{16}\Delta$ where $a = \frac{1}{2\gamma}\int |S_2|^2\nu^2 d\mu$, $b = \frac{1}{2\gamma}\int |S_1|^2\nu^2 d\mu$, $d = \frac{1}{2\gamma}\int \Re(S_1^* S_2)\mu d\mu$, and $\Delta = \sqrt{[5(a+b) - 4(1-d)]^2 + 24(a-b)^2}$, dependent solely on the scattering matrix of the particle. The iteration equation (6) is solved using the similarity transformation of G and the autocorrelation $\langle |E_x^{(n)}|^2 \rangle = \langle \xi_1^{(n)} + \xi_5^{(n)} + \xi_7^{(n)} \rangle$ is obtained.

Other autocorrelations $\langle |E_{y,z}^{(n)}|^2 \rangle$ and $\langle (s_{x,y,z}^{(n)})^2 \rangle$ can be treated similarly and they are governed by the *same* transfer matrix G. We find

$$\langle |E_\alpha^{(n)}|^2 \rangle = \frac{1}{3} + f_\alpha^+ \lambda_+^n + f_\alpha^- \lambda_-^n, \tag{7}$$

where $f_1^\pm = \frac{1}{3} \mp \frac{1}{3\Delta}(2a - 7b - 2d + 2)$, $f_2^\pm = -\frac{1}{6} \mp \frac{1}{6\Delta}(13a + 7b + 8d - 8)$, and $f_3^\pm = -\frac{1}{6} \pm \frac{1}{6\Delta} \times (17a - 7b + 4d - 4)$. The autocorrelation of $\mathbf{s}^{(n)}$ should be weighted by light intensity $|E_1^{(n)}|^2 + |E_2^{(n)}|^2$ in the average over the whole solid angle. We find

$$\langle |s_\alpha^{(n)}|^2 \rangle = \frac{1}{3} + h_\alpha^+ \lambda_+^n + h_\alpha^- \lambda_-^n, \tag{8}$$

where $h_1^\pm = -\frac{1}{6} \pm \frac{1}{6\Delta}(11a - b + 4d - 4)$, $h_2^\pm = -\frac{1}{6} \mp \frac{1}{6\Delta}(a - 11b - 4d + 4)$, and $h_3^\pm = \frac{1}{3} \mp \frac{1}{3\Delta} \times (5a + 5b + 4d - 4)$. The coefficients $f_\alpha^\pm$ and $h_\alpha^\pm$ satisfy $\sum_\alpha f_\alpha^\pm = \sum_\alpha h_\alpha^\pm = 0$. All cross correlations $\langle E_\alpha^{(n)}(E_\beta^{(n)})^* \rangle$ and $\langle s_\alpha^{(n)} s_\beta^{(n)} \rangle$ when $\alpha \neq \beta$ vanish due to the structure (4) of C [23].

If the incident light of unit intensity is in a general (elliptic) polarization state, $E_1^{(0)}\hat{x} + E_2^{(0)}\hat{y}$, where $\hat{x}$ and $\hat{y}$ are the principal axes of the polarization ellipse and $|E_1^{(0)}|^2 + |E_2^{(0)}|^2 = 1$, Eqs. (7) and (8) still hold with the only adjustment of coefficients $f_\alpha^\pm$ and $h_\alpha^\pm$ for $\alpha = 1, 2$ to

$|E_1^{(0)}|^2 f_\alpha^\pm + |E_2^{(0)}|^2 f_{3-\alpha}^\pm$ and $|E_1^{(0)}|^2 h_\alpha^\pm + |E_2^{(0)}|^2 h_{3-\alpha}^\pm$, respectively. $\langle |E_z^{(n)}|^2\rangle$ and $\langle (s_z^{(n)})^2\rangle$ do not change with the state of polarization of the incident light.

The values of the two eigenvalues $\lambda_\pm$ as a function of the size parameter x of the particle are displayed in Fig. 1. It should be noted that $\lambda_+ = 0.7$ and $\lambda_- = 0$ when $x \to 0$, incorporating Rayleigh scattering [24] as a special case. When the particle departs from Rayleigh scattering, the eigenvalues $\lambda_\pm$ initially increase rapidly and then oscillates with x due to Mie resonance scattering.

Noting that $0 \le \lambda_- < \lambda_+ < 1$, Eqs. (7) and (8) describe the following picture of polarized light isotropization in turbid media. The light propagation direction gradually becomes isotropic and light intensity equalizes in all polarization directions with increase of the number n of scattering events, i.e., $\langle |E_\alpha^{(n)}|^2\rangle \to \frac{1}{3}$ and $\langle |s_\alpha^{(n)}|^2\rangle \to \frac{1}{3}$. The decay of both light polarization and directionality anisotropies follows a power law of the larger eigenvalue $\lambda_+^n = \lambda_+^{l/l_s} = e^{-l/l_p}$ when $n \gg 1/\ln\frac{\lambda_+}{\lambda_-}$ where l is the path length of light, l_s is the scattering mean free path, and the isotropization length $l_p \equiv l_s/\ln\frac{1}{\lambda_+}$ specifies the characteristic length of light becoming isotropic in its polarization and propagation directions. The same length scale l_p is responsible for both isotropization of light propagation and polarization directions and light linear depolarization. The depolarization observed in multiple scattering is the result of summation of the polarizations of rays propagating along different random paths. Light linear depolarization occurs simultaneously with the isotropization of the beam of light.

For an incident light polarized along the x axis and propagating in the positive z axis, the fraction of co- and cross-polarized light is given by

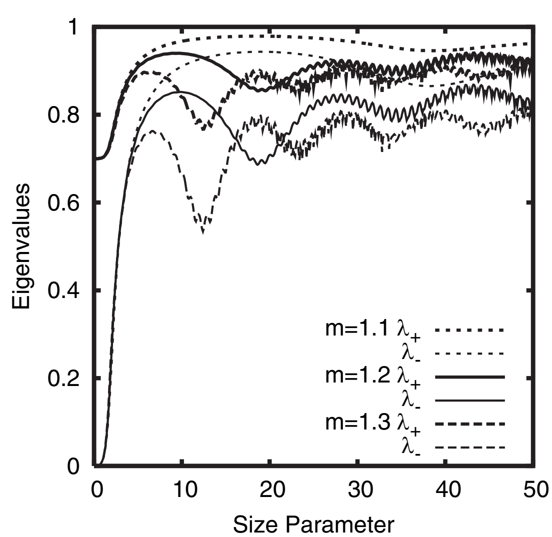

FIG. 1. The eigenvalues $\lambda_\pm$ versus the size parameter of Mie scatterers for various relative refractive index m. Size parameter is defined as $2\pi a/\lambda$ where a is the radius of the particle and λ is the wavelength of light in the background medium.

$$C_{\parallel,\perp}^{(n+1)} = \frac{\frac{1}{3} + f_{1,2}^+\lambda_+^n + f_{1,2}^-\lambda_-^n}{\frac{2}{3} - f_3^+\lambda_+^n - f_3^-\lambda_-^n} \simeq \frac{\frac{1}{3} + f_{1,2}^+ e^{-l/l_p}}{\frac{2}{3} - f_3^+ e^{-l/l_p}} \quad (9)$$

after $(n+1)$ scattering, where light is scattered to the z direction at the last scattering [25]. The second equality holds when $n \gg 1/\ln\frac{\lambda_+}{\lambda_-}$. l_p is seen to also characterize depolarization of incident linearly polarized light. Equation (9) reduces to the known result in [24] for Rayleigh scatterers whose $l_p = l_s/\ln\frac{10}{7} = 2.8 l_s$.

It is most instructive to contrast l_p with the transport mean free path l_t. l_p is the length scale at which light gets isotropized, while l_t is the distance over which the initial propagation direction of light gets forgotten and equals to $l_s/(1-g)$. The ratio l_p/l_t for Mie scatterers of various refractive indices and size parameters are displayed in Fig. 2. The complex oscillation structure in l_p/l_t originates from Mie resonance scattering.

Some observations can be made from Fig. 2. It shows $l_p > l_t$ as it is more difficult to achieve isotropization of propagation directions of light ($\langle s_{x,y,z}^2\rangle$ tends to 1/3) than to lose the memory of the initial propagation direction of light ($\langle \mathbf{s}\rangle$ tends to 0). Linearly polarized light is found to be, in general, harder to depolarize by smaller particles ($l_p \sim 2.8 l_t$) than by larger particles ($l_p \sim l_t - 1.5 l_t$) when light travels the same distance measured in l_t. But this rule is no longer true for particles of higher refractive indices [see Fig. 2(b)]. The ratio l_p/l_t is multiple valued for $g \sim 1$ as larger particles of different sizes may have the same g yet different l_p.

Surprisingly, for particles of a modest relative refractive index ($m < 1.3$), the inset in Fig. 2(a) demonstrates a simple linear relation between l_p/l_t and g, given by

$$l_p/l_t \simeq 1.8(1 - g/g_0) + 1 \simeq 2.8 - 2.5g \quad (10)$$

for particles of $g \le g_0 \sim 0.72 \pm 0.02$, where g_0 takes a slightly smaller value for larger m. Equation (10) agrees well with an empirical expression $l_p/l_t = 2.82 - 2.37g$ obtained from fitting DWS measurements and Monte Carlo simulations in Fig. (4a) of [8]. This relation (10) can be attributed to Rayleigh-Gans scattering of such particles [21] and extends the known result for Rayleigh scattering to a much larger regime.

We note the work reported here considers uncorrelated scatterers. The correlation effect in a strongly correlated colloidal suspension of small particles can be taken into account in the first order by introduction of a structure factor [26] and scaling the scattering matrix accordingly. The analysis shows Eq. (10) approximately holds in that regime for small particles. Correlation between scatterers increases the ratio l_p/l_t and may result in an abnormally slow linear depolarization of light. The backscattering cone [4,22] can be similarly treated using the model presented in this Letter.

In conclusion, we have presented a random walk model of vector photons to describe multiple scattering and de-

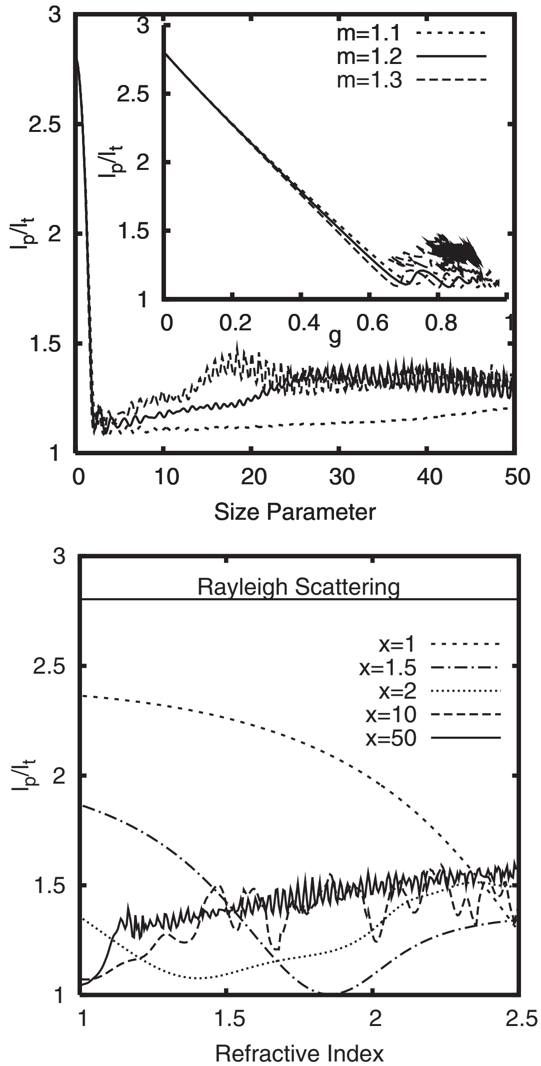

FIG. 2. The ratio of the characteristic length l_p of light becoming isotropic in its polarization and propagation directions over the transport mean free path l_t versus (a) size parameter x and (b) relative refractive index m. The inset in (a) shows $l_p/l_t \simeq 2.8 - 2.5g$ for particles with modest refractive index and average cosine $g \leq g_0 \sim 0.72$.

polarization of polarized light in turbid media. The isotropization of the polarization and propagation directions of polarized light has been shown to occur simultaneously and is governed by the same isotropization length. The isotropization length for Mie scatterers has been derived analytically, revealing the complex dependence of light isotropization on particle size and refractive index due to Mie resonance and interference structures. This work should facilitate quantitative understanding of isotropization and depolarization of multiply scattered light by Mie particles and randomly oriented nonspherical scatterers..

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[1] S. Chandrasekhar, *Radiative Transfer* (Dover, New York, 1960).
[2] P.-E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).
[3] M. P. V. Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).
[4] E. Akkermans, P. E. Wolf, and R. Maynard, Phys. Rev. Lett. **56**, 1471 (1986).
[5] M. P. van Albada, M. B. van der Mark, and A. Lagendijk, Phys. Rev. Lett. **58**, 361 (1987).
[6] D. J. Pine, D. A. Weitz, P. M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[7] F. C. MacKintosh and S. John, Phys. Rev. B **40**, 2383 (1989).
[8] L. F. Rojas-Ochoa, D. Lacoste, R. Lenke, P. Schurtenberger, and F. Scheffold, J. Opt. Soc. Am. A **21**, 1799 (2004).
[9] R. Carminati, R. Elaloufi, and J. J. Greffet, Phys. Rev. Lett. **92**, 213903 (2004).
[10] F. C. MacKintosh, J. X. Zhu, D. J. Pine, and D. A. Weitz, Phys. Rev. B **40**, R9342 (1989).
[11] K. M. Yoo and R. R. Alfano, Phys. Lett. A **142**, 531 (1989).
[12] D. Bicout, C. Brosseau, A. S. Martinez, and J. M. Schmitt, Phys. Rev. E **49**, 1767 (1994).
[13] A. D. Kim and M. Moscoso, Phys. Rev. E **64**, 026612 (2001).
[14] N. Ghosh, A. Pradhan, P. K. Gupta, S. Gupta, V. Jaiswal, and R. P. Singh, Phys. Rev. E **70**, 066607 (2004).
[15] M. J. Stephen and G. Cwilich, Phys. Rev. B **34**, 7564 (1986).
[16] D. Bicout and C. Brosseau, J. Phys. I (France) **2**, 2047 (1992).
[17] E. E. Gorodnichev, A. I. Kuzovlev, and D. B. Rogozkin, JETP **88**, 421 (1999).
[18] A. Yodh and B. Chance, Phys. Today **48**, No. 3, 38 (1995).
[19] M. C. W. van Rossum and T. M. Nieuwenhuizen, Rev. Mod. Phys. **71**, 313 (1999).
[20] M. Xu, Opt. Express **12**, 6530 (2004).
[21] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (John Wiley & Sons, New York, 1983).
[22] M. P. van Albada and A. Lagendijk, Phys. Rev. B **36**, R2353 (1987).
[23] Correlation $\langle M_{ij}^{(n)} (M_{kl}^{(n)})^* \rangle$ can be nonzero only when $j = l$, or $(j, l) = (1, 5)$, $(5, 1)$, $(2, 4)$, or $(4, 2)$ from recursion of Eq. (5).
[24] E. Akkermans, P. E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49**, 77 (1988).
[25] We have assumed the last scattering is Rayleigh-like ($\mathbf{E}^{(n+1)} = (\hat{z} \times \mathbf{E}^{(n)}) \times \hat{z}$) such that $E_{x,y}^{(n+1)} = E_{x,y}^{(n)}$. See also Refs. [4,22].
[26] P. M. Saulnier, M. P. Zinkin, and G. H. Watson, Phys. Rev. B **42**, R2621 (1990).