Diffusion coefficient depends on time, not on absorption

Wei Cai, M. Xu, Melvin Lax, and R. R. Alfano

Institute for Ultrafast Spectroscopy and Lasers, New York State Center of Advanced Technology for Ultrafast Photonic Materials and Applications, Department of Physics, The City College and Graduate Center of City University of New York, New York, New York 10031

Received November 27, 2001

The recent controversy over whether the photon diffusion coefficient depends on absorption is addressed by use of the analytical solution of the photon transport equation in an infinite homogeneous scattering medium. The diffusion coefficient is found to be independent of absorption but temporally dependent. After a long period of time, the photon diffusion coefficient approaches $D = 1/3\mu_s'$, which supports a claim made by Furutsu and Yamada [Phys. Rev. E **50**, 3634 (1994)]. At early times, the diffusion coefficient is smaller than $D = 1/3\mu_s'$, but this reduction cannot be expressed as $D = 1/3(\mu_s' + \mu_a)$, since the time-dependent diffusion coefficient is found to be unrelated to absorption.

OCIS codes: 290.1990, 030.5620, 170.5280, 290.7050.

The diffusion approximation of the radiative transfer equation has been used extensively in the study of photon migration and applications in turbid media. A controversy exists about the form of diffusion coefficient D in media in coexistence with scattering and absorption. In traditional derivations of the diffusion equation from the transport equation, the diffusion coefficient is obtained as[1–4]

$$D = 1/3(\mu_s' + \mu_a), \tag{1}$$

where μ_s' is the reduced scattering coefficient and μ_a is the absorption coefficient. Furutsu and Yamada[5] and Furutsu[6] pointed out that this form of D does not consist of the transformation property of the transport equation in an infinite homogeneous turbid medium: the photon distribution function at position $\mathbf{r}$, in direction $\mathbf{s}$ and time t, $I(\mathbf{r}, \mathbf{s}, t) = \exp(-c\mu_a t)I_0(\mathbf{r}, \mathbf{s}, t)$, where $I_0(\mathbf{r}, \mathbf{s}, t)$ is the solution for the same medium but without absorption. They[5,6] suggested another derivation that leads to

$$D = l_t/3 = 1/3\mu_s', \tag{2}$$

which is independent of absorption, where l_t is the transport mean-free path. This claim has been supported by Bassani *et al.*,[7] Durduran *et al.*,[8] and Nakai *et al.*[9] based on experiments and Monte Carlo simulations. In contrast, in later papers Aronson and Corngold,[10] Rinzema *et al.*,[11] and Durian[12], based on their experiments and numerical simulations, asserted that D should depend on absorption.

Most recently, we developed an analytical solution of the transport equation in an infinite homogeneous medium.[13,14] We derived[14] an expression for the spatial cumulants of the photon distribution function $I(\mathbf{r}, \mathbf{s}, t)$ at any direction $\mathbf{s}$ and time t, exact up to an arbitrarily high cumulant order, which can be used for accurate computation of the photon distribution function. Up to the second cumulant order, we obtained an approximate Gaussian spatial distribution that has the exact central position and the exact half-width of the distribution.[13] For photon density $N(\mathbf{r}, t)$, at $t \to \infty$ our result approaches the center-moved diffusion approximation (CMDA), with $D = 1/3\mu_s'$ independent of absorption. At a finite time, the diffusion coefficient D increased from zero at $t = 0$ to the above value at $t \to \infty$. Typically, in the case of the anisotropic factor $g \sim 0.9$ it takes approximately $10l_t$ for the diffusion mode to be valid, as shown experimentally by Yoo *et al.*[15] This $D(t)$ as a function of time is determined only by scattering parameters unrelated to absorption. Hence, it is physically unreasonable to use Eq. (1) to reduce the value of D at a finite time to fit experimental or simulated data.

The time-dependent photon density $N(\mathbf{r}, t)$ obtained from the CMDA in an infinite homogeneous medium for a collimated pulse source located at $\mathbf{r} = 0$ with incident direction along $\hat{\mathbf{z}}$ is given by[16]

$$N(\mathbf{r}, t) = \frac{1}{(4\pi Dct)^{3/2}} \exp\left[-\frac{(\mathbf{r} - l_t\hat{\mathbf{z}})^2}{4Dct} - \mu_a ct\right], \tag{3}$$

where D is the diffusion constant. This solution differs from the standard diffusion solution in that the center of the distribution is moved by $1l_t$ from the source position along the incident direction. On the other hand, our cumulant approximation (CUMA) for photon density, exact up to the second-order cumulant from the same source, is obtained as[13,14]

$$N(\mathbf{r}, t) = \frac{1}{(4\pi D_{zz}ct)^{1/2}} \frac{1}{4\pi D_{xx}ct} \exp\left[-\frac{(z - R_z)^2}{4D_{zz}ct}\right] \times \exp\left[-\frac{(x^2 + y^2)}{4D_{xx}ct}\right]\exp(-\mu_a ct), \tag{4}$$

with the moving center located at

$$R_z = c[1 - \exp(-g_1 t)]/g_1. \tag{5}$$

The corresponding diffusion coefficients are given by

0146-9592/02/090731-03$15.00/0

$$D_{zz} = \frac{c}{3t}\left\{\frac{t}{g_1} - \frac{3g_1 - g_2}{g_1{}^2(g_1 - g_2)}[1 - \exp(-g_1 t)] + \frac{2}{g_2(g_1 - g_2)}[1 - \exp(-g_2 t)] - \frac{3}{2g_1{}^2}[1 - \exp(-g_1 t)]^2\right\}, \quad (6)$$

$$D_{xx} = D_{yy} = \frac{c}{3t}\left\{\frac{t}{g_1} + \frac{g_2}{g_1{}^2(g_1 - g_2)}[1 - \exp(-g_1 t)] - \frac{1}{g_2(g_1 - g_2)}[1 - \exp(-g_2 t)]\right\}. \quad (7)$$

Here $g_l = \mu_s c[1 - a_l/(2l + 1)]$, where the single-scattering phase function is expanded in Legendre polynomials by

$$P(\cos\theta) = [1/(4\pi)]\sum_l a_l P_l(\cos\theta).$$

Two special values of g_l are $g_0 = 0$ and $g_1 = c/l_t$.

The original meaning of diffusion is a description of a random process. As early as when Brownian motion was first studied, researchers have known that under random forces from the surrounding medium particles will take a diffusion process, namely, spread from the center outward. The diffusion coefficient is a parameter that characterizes the rate of spread, which can be time dependent. The standard diffusion equation with a diffusion constant is the simplest form with which to describe these phenomena. The photon propagation in a turbid medium is one example of the random processes, which is more complicated than that described by the standard diffusion equation, mainly because the photons are injected with velocity along a direction. This leads to photon propagation from ballistic to snakelike and then to the diffusive mode. The diffusion coefficient in this process should be time dependent.

The fundamental time-independent parameters of the medium are the scattering coefficients, the absorption coefficients, and the phase function in the radiative transfer equation, because they have definite meaning from a microscopic viewpoint. When regarding the diffusion constant as a physical parameter, we should note that this concept is not original but is a secondary quantity that is derived by use of an approximate method. Of course, these two meanings of a diffusion coefficient are not incompatible. When a standard diffusion equation is approximately valid, the diffusion constant is correct according to both meanings.

Although the Gaussian distribution in Eq. (4) is an approximation because it cuts into the second-order cumulant, Eqs. (6) and (7) provide an exact description of the half-width of the real distribution.

Figure 1 shows the diffusion coefficients from CUMA, D_{zz} and D_{xx} [Eqs. (6) and (7)], as a function of time, where g_l are calculated by Mie theory[17] assuming (for this figure) that the homogeneous scattering medium consists of water droplets with $\rho/\lambda = 1$ uniformly distributed in air, with ρ the radius of the droplet, λ the wavelength of light, and index of refraction $m = 1.33$. The anisotropic factor for this case is $a_1/3 = 0.8436$.

At $t \to \infty$, the photon density in Eq. (4) approaches Eq. (3), where the diffusion coefficients D_{zz} and D_{xx} approach $D = 1/3\mu_s'$, not Eq. (1). This means that Eq. (1) for a diffusion coefficient that depends on absorption leads to an incorrect result even for an infinite time limit. On the other hand, the central-limit theorem claims that the obtained Gaussian distribution would be accurate after a large number of collisions. Hence, Eq. (3) with $D = 1/3\mu_s'$ provides an accurate solution of photon transport in an infinite homogeneous medium at an infinite time limit, no matter whether absorption is strong or weak. At finite times, the diffusion coefficient is always smaller than its value at infinite time. In fact, at early time $t \to 0$, the center moves as $ct\hat{\mathbf{z}}$ and the diffusion coefficient approaches zero. This result presents a clear picture of nearly ballistic motion at $t \to 0$. With an increase in time, the motion at the center slows down, and the diffusion coefficient increases from zero. This stage of photon migration is in the snakelike mode. For a large period of time, Eq. (4) approaches the center-moved diffusion solution. As shown in Eqs. (6) and (7), the time-dependent diffusion coefficient is determined only by the scattering parameters g_l and is unrelated to the absorption coefficient μ_a. Hence, to fit data of numerical simulation or experiments at finite time, one should not use Eq. (1) to reduce the diffusion coefficient.

The Aronson and Corngold paper[10] emphasizes that the time-independent diffusion equation rather than the time-dependent equation should be examined to determine which form of diffusion coefficient is correct. The time-independent solution can be obtained by integration of the time-dependent solution over time. Integration of Eq. (3) over time yields the time-independent diffusive solution in an infinite homogeneous medium:

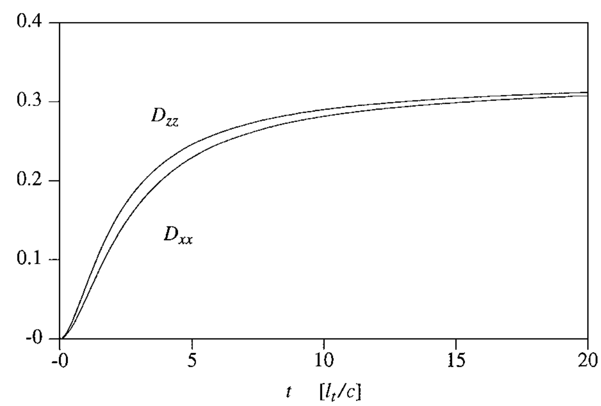

Fig. 1. Diffusion coefficients D_{zz} and D_{xx} from Eqs. (6) and (7) as a function of time t.

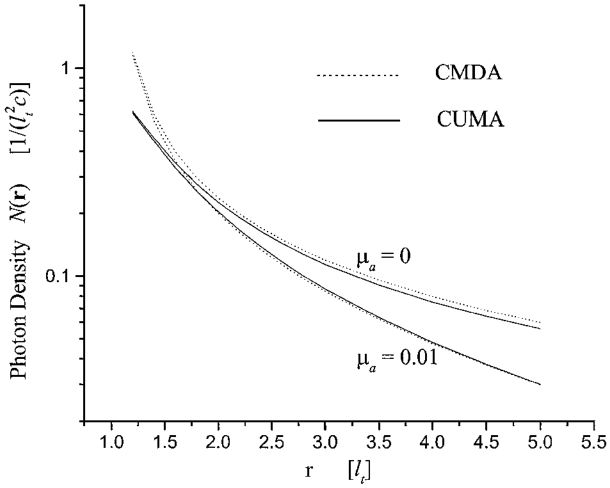

Fig. 2. Steady-state photon density as a function of distance from the source (along the incident direction) for different absorption coefficients μ_a with unit $1/l_t$ obtained with the CMDA and the CUMA. The unit of length is l_t; the unit of time is l_t/c.

$$N(\mathbf{r}) = \frac{1}{4\pi Dc|\mathbf{r} - l_t\hat{\mathbf{z}}|}\exp[-|\mathbf{r} - l_t\hat{\mathbf{z}}|(\mu_a/D)^{1/2}]. \quad (8)$$

A more accurate steady-state distribution $N(\mathbf{r})$ in an infinite homogeneous medium can be obtained by integration of Eq. (4) over time from $t = 0$ to $t = \infty$. Figure 2 shows $N(\mathbf{r})$ as a function of r with the detector set at (0, 0, r) for different absorption coefficients μ_a. The dashed curves were obtained from Eq. (8) with $D = l_t/3$, and the solid curves were obtained by integration of our time-dependent cumulant solution in Eq. (4) over time, with the corresponding diffusion coefficients given by Eqs. (6) and (7). When the detector is near the source, the photon density from CUMA is distinctly smaller than that from Eq. (8). This happens because early time photons play an important role, and the diffusion approximation fails when it is near the source. With an increase in distance between source and detector, the results from the CMDA and CUMA approaches are in agreement for different absorption coefficients. These results confirm that the diffusion coefficient should be $D = l_t/3$ at the diffusion limit, independently of absorption.

In conclusion, our analytical solution of the time-dependent photon density in an infinite homogeneous medium supports the claim that the diffusion coefficients is independent of absorption but is temporally dependent.

This research is supported in part by NASA Institutional Research Awards, U.S. Army Medical Research and Materiels Command, the U.S. Army Research Office, the U.S. Department of Energy, and the Defense Advanced Research Projects Agency.

References

1. A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic, New York, 1978), Vol. 1.
2. S. Glasstone and M. C. Edlund, *The Elements of Nuclear Reactor Theory* (Van Nostrand, New York, 1952).
3. W. M. Star, J. P. Marijnissen, and M. J. C. van Gemert, Phys. Med. Biol. **33,** 437 (1988).
4. J. J. Duderstadt and W. R. Martin, *Transport Theory* (Wiley, New York, 1979).
5. K. Furutsu and Y. Yamada, Phys. Rev. E **50,** 3634 (1994).
6. K. Furutsu, J. Opt. Soc. Am. A **14,** 267 (1997).
7. M. Bassani, F. Martelli, G. Zaccanti, and D. Contini, Opt. Lett. **22,** 853 (1997).
8. T. Durduran, A. G. Yodh, B. Chance, and D. A. Boas, J. Opt. Soc. Am. A **14,** 3358 (1997).
9. T. Nakai, G. Nishimura, K. Yamamoto, and M. Tamura, Phys. Med. Biol. **42,** 2541 (1997).
10. R. Aronson and N. Corngold, J. Opt. Soc. Am. A **16,** 1066 (1999).
11. K. Rinzema, L. H. P. Murrer, and W. M. Star, J. Opt. Soc. Am. A **15,** 2078 (1998).
12. D. J. Durian, Opt. Lett. **23,** 1502 (1998).
13. W. Cai, M. Lax, and R. R. Alfano, Phys. Rev. E **61,** 3871 (2000).
14. W. Cai, M. Lax, and R. R. Alfano, J. Phys. Chem. B **104,** 3996 (2000).
15. K. M. Yoo, F. Liu, and R. R. Alfano, Phys. Rev. Lett. **64,** 2647 (1990).
16. M. S. Patterson, B. Chance, and B. C. Wilson, Appl. Opt. **28,** 2331 (1995).
17. C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 1983).