Analytical solutions for the mean-square displacement derived from transport theory

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In this paper, exact analytical solutions are derived for the second-order moment and the mean-square displacement based on the transport theory fluence that is caused by an arbitrary anisotropic point source, which is located at the origin of a three-dimensional coordinate system. In particular, the derivations are carried out as a function of the number of scattering events. The resulting formulas in the steady-state and time domain depend, apart from the scattering coefficient and partly the absorption coefficient, only on the anisotropy factor of the considered rotationally invariant scattering phase function. We additionally present the second-order moment and the mean-square displacement for the fluence of fluorescence light in the steady-state domain.

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I. INTRODUCTION

Studies of the mean-square displacement (MSD) are prevalent in many different scientific areas, such as in heat conduction [1], in ballistic and diffusive Brownian motion [2], in light transport in clouds [3] or in glaciers [4], in transport in disordered media [5], in animal movement [6], in quantum walks [7], in superdiffusion and subdiffusion [8], and in further stochastic problems in physics, chemistry, and electrical engineering [9].

The MSD of particles in an infinitely extended random medium has been studied for more than 100 years. For example, Einstein derived his famous formula in the time domain investigating Brownian motion [10]. He applied the diffusion theory and found that $\langle r^2(t)\rangle$ is proportional to t, which is valid only for long time values as he already noted. Shortly afterwards, Langevin obtained a solution based on Newton's second law, assuming a random (isotropic) force. This formula exhibited the same long time behavior, but was applicable also for early time values [11]. Later, a solution of the telegrapher's equation, an approximation of the more fundamental transport equation, was derived, which delivered the same solution as was obtained by Langevin [12]. Further, random walk models were applied to obtain the formula for the MSD for anisotropic "turn angles" [13]. Investigations of the MSD in the steady-state domain were performed especially in optics, where $\langle r^2\rangle$ is finite due to the fact that photons have a nonzero probability of being absorbed when propagating through random media. Formulas were derived, both, based on the radiative transport theory [14,15] and on the diffusion theory [16], which is a low-order approximation to the radiative transport equation (RTE).

In this study, we derive the second-order moment and the MSD for the fluence [17] based on the exact transport theory in the steady state and time domain that undergoes an arbitrary number of scattering events. In this context, the radiance is expanded in terms of successive scattering orders, which results, in view of the second-order moment, in a second-order linear difference equation. The corresponding moments in time domain are obtained under consideration of the Laplace transform. The resulting formulas are valid for all anisotropic point sources and an arbitrary rotationally invariant scattering phase function. We note that the derived equations are exact within the RTE, e.g., for arbitrary time values or any desired scattering-to-absorption ratio. Thus, they provide a detailed information on the characteristics of photon transport for any regime of propagation. The derived expressions are verified and illustrated by comparisons with the Monte Carlo (MC) method.

II. SECOND-ORDER MOMENT AND MSD FOR THE TRANSPORT THEORY FLUENCE

The starting point of the derivations is the three-dimensional RTE for an anisotropic point source that is given by

$$\begin{aligned}\boldsymbol{\Omega}\cdot\nabla I(\mathbf{x},\boldsymbol{\Omega})+\mu_t I(\mathbf{x},\boldsymbol{\Omega})\\ =\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')I(\mathbf{x},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'+\delta(\mathbf{x})Q(\boldsymbol{\Omega}),\end{aligned}\tag{1}$$

where $(\mathbf{x},\boldsymbol{\Omega})\in\mathbb{R}^3\times\mathbb{S}^2$, I is the radiance, $\Phi(\mathbf{x}):=\int_{\mathbb{S}^2} I(\mathbf{x},\boldsymbol{\Omega})\,d\boldsymbol{\Omega}$ denotes the fluence, f is the scattering phase function that is normalized according to $\int f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'=1$, $\mu_t=\mu_a+\mu_s$ is the total attenuation coefficient, μ_a the absorption coefficient, and μ_s the scattering coefficient. Concerning the source term, we have the normalization $\int Q(\boldsymbol{\Omega})\,d\boldsymbol{\Omega}=1$. In the following, we derive an expression for the second-order moment of the fluence in dependence of the number of scattering events. For this task, the radiance of Eq. (1) is formally expanded in terms of successive scattering orders according to the Neumann series [18,19]

$$I(\mathbf{x},\boldsymbol{\Omega})=\sum_{n=0}^{\infty} I_n(\mathbf{x},\boldsymbol{\Omega}),$$

with $(I_n)_{n\geqslant 0}$ being a sequence of functions whose terms are governed by the following recursively defined transport equation [19,20]:

$$\boldsymbol{\Omega}\cdot\nabla I_n(\mathbf{x},\boldsymbol{\Omega})+\mu_t I_n(\mathbf{x},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')I_{n-1}(\mathbf{x},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}',\quad n=1,2,\ldots, \tag{2}$$

where the initial term I_0 is given as solution of

$$\boldsymbol{\Omega}\cdot\nabla I_0(\mathbf{x},\boldsymbol{\Omega})+\mu_t I_0(\mathbf{x},\boldsymbol{\Omega})=\delta(\mathbf{x})Q(\boldsymbol{\Omega})\quad\Longrightarrow\quad I_0(\mathbf{x},\boldsymbol{\Omega})=\frac{e^{-\mu_t r}}{r^2}Q(\boldsymbol{\Omega})\delta(\hat{\mathbf{x}}-\boldsymbol{\Omega}),$$

with $r=\|\mathbf{x}\|$ and $\hat{\mathbf{x}}=\mathbf{x}/r$. The fluence in terms of scattering orders is $\Phi(\mathbf{x})=\sum_{n=0}^{\infty}\Phi_n(\mathbf{x})$. The second-order moment $\langle x^2\rangle$ of a function $\varphi:\mathbb{R}^3\to\mathbb{R}$ is formally defined as

$$\langle x^2\rangle=\int_{\mathbb{R}^3} r^2\varphi(\mathbf{x})\,d\mathbf{x}=-\Delta_k\int_{\mathbb{R}^3}\varphi(\mathbf{x})\exp(-i\mathbf{k}\cdot\mathbf{x})\,d\mathbf{x}\bigg|_{\mathbf{k}=\mathbf{0}}=-\Delta_k\widehat{\varphi}(\mathbf{0}),$$

with $\widehat{\varphi}(\mathbf{k})=\int_{\mathbb{R}^3}\varphi(\mathbf{x})\exp(-i\mathbf{k}\cdot\mathbf{x})\,d\mathbf{x}$ being the corresponding Fourier transform (FT). Below, we neglect the index k and write Δ instead of Δ_k. To derive the second-order moment for the fluence after $n\in\mathbb{N}_0$ scattering events, we start with Eq. (2) in Fourier space that is given by

$$\mu_t\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})+i\mathbf{k}\cdot\boldsymbol{\Omega}\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\widehat{I}_{n-1}(\mathbf{k},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'. \tag{3}$$

Applying the Laplace operator on both sides of Eq. (3) under consideration of the product rule $\Delta[(i\mathbf{k}\cdot\boldsymbol{\Omega})\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})]=(i\mathbf{k}\cdot\boldsymbol{\Omega})\Delta\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})+2i\,\boldsymbol{\Omega}\cdot\nabla\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})$ and setting $\mathbf{k}=\mathbf{0}$ gives

$$\mu_t\Delta\widehat{I_n}(\mathbf{0},\boldsymbol{\Omega})+2i\boldsymbol{\Omega}\cdot\nabla\widehat{I_n}(\mathbf{0},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\Delta\widehat{I}_{n-1}(\mathbf{0},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'.$$

Integrating this equation over the unit sphere $\mathbb{S}^2$ results in

$$\mu_t\Delta\widehat{\Phi}_n(\mathbf{0})+2i\,\mathrm{div}\,\widehat{\mathbf{J}}_n(\mathbf{0})=\mu_s\Delta\widehat{\Phi}_{n-1}(\mathbf{0}), \tag{4}$$

where $\widehat{\mathbf{J}}(\cdot):=\int_{\mathbb{S}^2}\boldsymbol{\Omega}\widehat{I}(\cdot,\boldsymbol{\Omega})\,d\boldsymbol{\Omega}$ denotes the flux vector. To eliminate the quantity $\mathrm{div}\,\widehat{\mathbf{J}}_n(\mathbf{0})$ a second relation is needed. The application of the Nabla operator on both sides of Eq. (3) gives

$$(\mu_t+i\mathbf{k}\cdot\boldsymbol{\Omega})\nabla\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})+i\boldsymbol{\Omega}\widehat{I_n}(\mathbf{k},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\nabla\widehat{I}_{n-1}(\mathbf{k},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'.$$

Setting $\mathbf{k}=\mathbf{0}$ and multiplying both sides with the unit vector $\boldsymbol{\Omega}$ leads to

$$\mu_t\,\boldsymbol{\Omega}\cdot\nabla\widehat{I_n}(\mathbf{0},\boldsymbol{\Omega})+i\widehat{I_n}(\mathbf{0},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2}\boldsymbol{\Omega}f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\nabla\widehat{I}_{n-1}(\mathbf{0},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'.$$

Integrating this equation over the unit sphere $\mathbb{S}^2$ results in the continuity equation

$$\mu_t\,\mathrm{div}\,\widehat{\mathbf{J}}_n(\mathbf{0})+i\widehat{\Phi}_n(\mathbf{0})=\mu_s g\,\mathrm{div}\,\widehat{\mathbf{J}}_{n-1}(\mathbf{0}), \tag{5}$$

where we used $\int_{\mathbb{S}^2}\boldsymbol{\Omega}f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}=g\,\boldsymbol{\Omega}'$ with $|g|\leqslant 1$ being the anisotropy factor of the phase function. To proceed further, we introduce $X_n:=-\Delta\widehat{\Phi}_n(\mathbf{0})=\int_{\mathbb{R}^3}x^2\Phi_n(\mathbf{x})\,d\mathbf{x}$, $Y_n:=\mathrm{div}\,\widehat{\mathbf{J}}_n(\mathbf{0})$, and $M_n:=\widehat{\Phi}_n(\mathbf{0})=\int_{\mathbb{R}^3}\Phi_n(\mathbf{x})\,d\mathbf{x}$, where X_n is the desired second-order moment. Equations (4) and (5) can then be written in form of the following system of linear difference equations:

$$\mu_t X_n-\mu_s X_{n-1}=2iY_n, \tag{6}$$

$$\mu_t Y_n-\mu_s g Y_{n-1}=-iM_n. \tag{7}$$

This system is reducible to a single second-order equation by eliminating Y_n. From Eq. (6) we get $Y_n=(\mu_t X_n-\mu_s X_{n-1})/(2i)$ and hence $Y_{n-1}=(\mu_t X_{n-1}-\mu_s X_{n-2})/(2i)$. Inserting these quantities into Eq. (7) results in

$$\mu_t^2 X_n-\mu_s\mu_t(1+g)X_{n-1}+\mu_s^2 gX_{n-2}=2M_n,\quad n=0,1,\ldots, \tag{8}$$

under consideration of $X_{-2}=X_{-1}=0$. The zero-order moment M_n can be found from Eq. (3). By setting $\mathbf{k}=\mathbf{0}$, we have

$$\mu_t\widehat{I_n}(\mathbf{0},\boldsymbol{\Omega})=\mu_s\int_{\mathbb{S}^2} f(\boldsymbol{\Omega}\cdot\boldsymbol{\Omega}')\widehat{I}_{n-1}(\mathbf{0},\boldsymbol{\Omega}')\,d\boldsymbol{\Omega}'.$$

Integration over the unit sphere $\mathbb{S}^2$ results in the recurrence relation $\mu_t M_n=\mu_s M_{n-1}$ subject to the initial value

$$M_0=\int_{\mathbb{R}^3}\Phi_0(\mathbf{x})\,d\mathbf{x}=\int_{\mathbb{R}^3}\frac{e^{-\mu_t r}}{r^2}Q(\hat{\mathbf{x}})\,d\mathbf{x}=\frac{1}{\mu_t}. \tag{9}$$

As a result, we find successively $M_1=\mu_s/\mu_t^2$, $M_2=\mu_s^2/\mu_t^3$, …, and $M_n=\mu_s^n/\mu_t^{n+1}$ for the zero-order moment. Furthermore, we need two initial conditions for solving Eq. (8). Setting $n=0$ and $n=1$ leads to the conditions $\mu_t^2X_0=2/\mu_t$ and $\mu_t^2X_1-\mu_s\mu_t(1+g)X_0=2\mu_s/\mu_t^2$ yielding $X_0=$

$2/\mu_t^3$ and $X_1 = 2\mu_s(2+g)/\mu_t^4$. Equation (8) can be solved similarly to an ordinary differential equation. To find the corresponding homogenous solution, we perform the ansatz $X_n^{(h)} := \lambda^n$. Inserting this into Eq. (8) gives the characteristic equation $\mu_t^2\lambda^2 - \mu_s\mu_t(1+g)\lambda + \mu_s^2 g = 0$ with the roots $\lambda_1 = \mu_s/\mu_t$ and $\lambda_2 = \mu_s g/\mu_t$. The particular part can be obtained via a similarity ansatz. In this context, we have to note that the right-hand side of Eq. (8) contains a part of the homogeneous solution, namely $M_n = \mu_s^n/\mu_t^{n+1} = \lambda_1^n/\mu_t$. Thus, instead of $X_n^{(p)} = K\lambda_1^n$, we have to use the modified ansatz $X_n^{(p)} = Kn\lambda_1^n$. Inserting this into Eq. (8) leads to the coefficient $K = 2/[\mu_t^3(1-g)]$. The complete solution of Eq. (8) in terms of the homogenous and particular part is given by

$$X_n = C_1\lambda_1^n + C_2\lambda_2^n + \frac{2}{\mu_t^3}\frac{n}{1-g}\left(\frac{\mu_s}{\mu_t}\right)^n, \quad n \in \mathbb{N}_0, \quad C_1, C_2 \in \mathbb{R}. \tag{10}$$

We note that this expression is formally not defined for $g = 1$. However, this special case can be obtained from the final solution in the form of a limit, see below. The unknown coefficients are fixed by the initial values X_0 and X_1, which leads to the following conditions:

$$C_1 + C_2 = \frac{2}{\mu_t^3} \quad \text{and} \quad \lambda_1 C_1 + \lambda_2 C_2 = \frac{2\mu_s}{\mu_t^4}\left[1 - \frac{g^2}{1-g}\right]. \tag{11}$$

The corresponding values are obtained as

$$C_1 = \frac{2}{\mu_t^3}\left[1 - \frac{g^2}{(1-g)^2}\right] \quad \text{and} \quad C_2 = \frac{2}{\mu_t^3}\frac{g^2}{(1-g)^2}. \tag{12}$$

The desired solution of Eq. (8) (the second-order moment to the transport theory fluence) can then be written as

$$X_n = \frac{2}{\mu_t^3}\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right]\left(\frac{\mu_s}{\mu_t}\right)^n, \quad n \in \mathbb{N}_0, \quad -1 \leqslant g < 1. \tag{13}$$

As mentioned above, the case $g = 1$ can be evaluated via ℓ'Hospital's rule applied to the limit

$$\begin{aligned}\lim_{g\to 1}&\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right]\\ &= \lim_{g\to 1}\frac{(1-g)^2 + (1-g)n - g^2(1-g^n)}{(1-g)^2}\\ &\overset{\ell'H}{=} \lim_{g\to 1}\frac{2(g-1) - n - 2g + (n+2)g^{n+1}}{2(g-1)}\\ &\overset{\ell'H}{=} \lim_{g\to 1}\frac{(n+2)(n+1)g^n}{2} = \frac{(n+1)(n+2)}{2},\end{aligned}$$

which results in $X_n = (n+1)(n+2)\mu_s^n/\mu_t^{n+3}$. In addition, summing over all scattering orders results in

$$\sum_{n=0}^{\infty} X_n = \frac{2}{\mu_a^2(\mu_a + \mu_s')} < \infty, \quad \text{if} \quad \mu_a > 0, \tag{14}$$

where $\mu_s' = (1-g)\mu_s$. We now can define the MSD belonging to the nth scattering event in the form of a normalized second-order moment according to

$$\langle r^2\rangle_n := \frac{X_n}{M_n} = \begin{cases}\frac{2}{\mu_t^2}\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right], & g \in [-1, 1),\\ \frac{(n+1)(n+2)}{\mu_t^2}, & g = 1.\end{cases} \tag{15}$$

Equation (15) has already been derived, only for a nonabsorbing medium, by following an extrapolation procedure [14]. However, it must be stressed that in Ref. [14] a rigorous mathematical proof of this relation was not presented. In addition, we can give the corresponding second-order difference equation belonging to the MSD. By dividing both sides of Eq. (8) by the zero-order moment M_n results in the equation

$$\langle r^2\rangle_n - (1+g)\langle r^2\rangle_{n-1} + g\langle r^2\rangle_{n-2} = 2/\mu_t^2, \quad n = 0, 1, \ldots,$$

with $\langle r^2\rangle_{-2} = \langle r^2\rangle_{-1} = 0$. In this case, we have $\langle r^2\rangle_n^{(h)} \in \text{span}\{1, g^n\}$ for $g \in [-1, 1)$ and $\langle r^2\rangle_n^{(h)} \in \text{span}\{1, n\}$ when $g = 1$. This difference equation can be solved similarly as Eq. (8) leading to the MSD (15). The MSD after an infinite number of scattering events is given by the ratio

$$\langle r^2\rangle = \frac{\sum_{n=0}^{\infty} X_n}{\sum_{n=0}^{\infty} M_n} = \frac{2}{\mu_a(\mu_a + \mu_s')} \quad \text{for} \quad \mu_a > 0. \tag{16}$$

We note that Eq. (16) was already derived earlier based on transport theory for the special case of an iso-delta scattering phase function [15]. Further, we note that the MSD (16) also follows from the diffusion approximation [16]. In addition to the steady-state domain, we also want to provide the corresponding results in the time domain caused by the point source $\delta(\mathbf{x})Q(\mathbf{\Omega})\delta(t)$. To find the second-order moment and the MSD, we extended the results derived above to the temporal frequency domain [21] by replacing the real-valued absorption μ_a by the complex-valued quantity $\mu_a + s/c$, with c being the speed of light and $s \in \mathbb{C}$ denotes the Laplace transform variable occurring within the integral transform $\mathcal{L}\{f(t)\}(s) = \hat{f}(s) = \int_0^\infty f(t)e^{-st}\,dt$. The extension of Eq. (1) to the temporal frequency domain becomes

$$\begin{aligned}\mathbf{\Omega}\cdot\nabla\widehat{I}(\mathbf{x}, \mathbf{\Omega}, s) &+ (\mu_t + s/c)\widehat{I}(\mathbf{x}, \mathbf{\Omega}, s)\\ &= \mu_s\int_{\mathbb{S}^2} f(\mathbf{\Omega}\cdot\mathbf{\Omega}')\widehat{I}(\mathbf{x}, \mathbf{\Omega}', s)\,d\mathbf{\Omega}' + \delta(\mathbf{x})Q(\mathbf{\Omega}).\end{aligned} \tag{17}$$

The second-order moment $\widehat{X}_n(s)$ belonging to the fluence $\widehat{\Phi}_n(\mathbf{x}, s)$ in Laplace space can be directly obtained from the previous section because the results in the steady-state domain are also valid for a complex-valued absorption. Therefore, we have the representation

$$\widehat{X}_n(s) = \begin{cases}\frac{2\mu_s^n}{(\mu_t+s/c)^{n+3}}\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right], & g \in [-1, 1),\\ (n+1)(n+2)\frac{\mu_s^n}{(\mu_t+s/c)^{n+3}}, & g = 1.\end{cases} \tag{18}$$

To invert the Laplace transform we note on the relation $\mathcal{L}^{-1}\{n!/(s+a)^{n+1}\}(t) = t^n e^{-at}$ [22], yielding in the time domain

$$X_n(t) = \begin{cases} \frac{2c\mu_s^n}{(n+2)!}\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right](ct)^{n+2}e^{-\mu_t ct}, & g \in [-1, 1), \\ c\mu_s^n\frac{(ct)^{n+2}}{n!}e^{-\mu_t ct}, & g = 1. \end{cases} \tag{19}$$

The summation with respect to the scattering order gives

$$\sum_{n=0}^{\infty} X_n(t) = \begin{cases} 2c\frac{\mu_s' ct - 1 + e^{-\mu_s' ct}}{(\mu_s')^2}e^{-\mu_a ct}, & g \in [-1, 1), \\ c(ct)^2 e^{-\mu_a ct}, & g = 1. \end{cases}$$

The time-dependent MDS for the nth scattering event is obtained via normalization according to

$$\langle r^2(t)\rangle_n := \frac{X_n(t)}{M_n(t)} = \begin{cases} \frac{2(ct)^2}{(n+1)(n+2)}\left[1 + \frac{n}{1-g} - g^2\frac{1-g^n}{(1-g)^2}\right], & g \in [-1, 1), \\ (ct)^2, & g = 1, \end{cases} \tag{20}$$

where we used $M_n(t) = \mathcal{L}^{-1}\{\mu_s^n/(\mu_t + s/c)^{n+1}\}(t) = c\mu_s^n(ct)^n e^{-\mu_t ct}/n!$. The MSD after an infinity number of scattering events becomes

$$\langle r^2(t)\rangle = \frac{\sum_{n=0}^{\infty} X_n(t)}{\sum_{n=0}^{\infty} M_n(t)} = \begin{cases} 2\frac{\mu_s' ct - 1 + e^{-\mu_s' ct}}{(\mu_s')^2}, & g \in [-1, 1), \\ (ct)^2, & g = 1. \end{cases} \tag{21}$$

In the case of $g \in [-1, 1)$, one obtains $\langle r^2(t)\rangle \approx 2ct/\mu_s'$ as $t \gg \mu_s' c$, which is in accordance with the well-known linear relationship from the diffusion theory [10]. For small time values $t \ll \mu_s' c$ one obtains $\langle r^2(t)\rangle \approx (ct)^2$ as expected for the ballistic regime. We note that Eq. (21) for $g = 0$ was already derived by Langevin for the special case of a random force [11], which corresponds to isotropic scattering in the framework of radiative transport theory.

In view of applications, another important aspect is the consideration of fluorescent phenomena. The associated RTE for describing the fluorescence photon migration in Fourier space is given by

$$\mu_{te}\widehat{I}_e(\mathbf{k}, \mathbf{\Omega}) + i\mathbf{k}\cdot\mathbf{\Omega}\widehat{I}_e(\mathbf{k}, \mathbf{\Omega}) = \mu_{se}\int_{\mathbb{S}^2} f_e(\mathbf{\Omega}\cdot\mathbf{\Omega}')\widehat{I}_e(\mathbf{k}, \mathbf{\Omega}')\, d\mathbf{\Omega}' + \frac{\mu_a}{4\pi}\widehat{\Phi}(\mathbf{k}), \tag{22}$$

where the subscript e refers to the emission wavelength and $\hat{\Phi}$ is the transformed fluence at the excitation wavelength caused by the anisotropic point source of Eq. (1). The corresponding second-order moment and the MSD can be derived in a similar way as shown above. The application of the Laplace operator on both sides of Eq. (22) leads for $\mathbf{k} = \mathbf{0}$ and after the integration over the unit sphere to

$$\mu_{ae}\Delta\widehat{\Phi}_e(\mathbf{0}) + 2i\,\mathrm{div}\,\widehat{\mathbf{J}}_e(\mathbf{0}) = \mu_a\Delta\widehat{\Phi}(\mathbf{0}). \tag{23}$$

In accordance with the derivations outlined above, we can deduce the following continuity equation at the emission wavelength:

$$\mu_{te}'\,\mathrm{div}\,\widehat{\mathbf{J}}_e(\mathbf{0}) + i\widehat{\Phi}_e(\mathbf{0}) = \frac{\mu_a}{4\pi}\int_{\mathbb{S}^2}\mathbf{\Omega}\cdot\nabla\widehat{\Phi}(\mathbf{0})\, d\mathbf{\Omega} = 0,$$

where $\mu_{te}' = \mu_{ae} + (1 - g_e)\mu_{se}$ and $g_e \in [-1, 1]$ being the anisotropy factor of the normalized phase function f_e. From Eq. (22), we find $\widehat{\Phi}_e(\mathbf{0}) = \mu_a\widehat{\Phi}(\mathbf{0})/\mu_{ae} = 1/\mu_{ae}$. In addition, the second-order moment $X = -\Delta\widehat{\Phi}(\mathbf{0}) = 2/(\mu_a^2\mu_t')$ is known from Eq. (14). Defining $X_e := -\Delta\widehat{\Phi}_e(\mathbf{0})$ and inserting $\mathrm{div}\,\widehat{\mathbf{J}}_e(\mathbf{0}) = -i/(\mu_{ae}\mu_{te}')$ into Eq. (23) results in

$$X_e = \frac{2}{\mu_a\mu_{ae}[\mu_a + \mu_s(1-g)]} + \frac{2}{\mu_{ae}^2[\mu_{ae} + \mu_{se}(1-g_e)]}. \tag{24}$$

The corresponding MSD at the excitation wavelength follows via normalization under the use of $M_e := 1/\mu_{ae}$, yielding

$$\langle r^2\rangle_e := \frac{X_e}{M_e} = \frac{2}{\mu_a[\mu_a + \mu_s(1-g)]} + \frac{2}{\mu_{ae}[\mu_{ae} + \mu_{se}(1-g_e)]}.$$

This result was proved for the restricted case of the iso-delta scattering phase function [15]. In addition, the second-order moment (24) can be extended to the temporal frequency domain to reconstruct the time-resolved MSD.

III. ILLUSTRATION AND VERIFICATION

In this section, the derived formulas for the second-order moment and the MSD are verified and illustrated by comparisons with a home-made Monte Carlo code [15]. The Monte Carlo method simulates the photons' paths through the scattering and absorbing medium and delivers in the limit of an infinitely large number of simulated photons exact solutions of the radiative transport theory. We start with the MSD in the steady-state domain that is given by Eq. (15). Figure 1 displays the analytical solution (solid line) and the data from the MC method (open circles) as a function of the scattering order. The optical properties are set to $\mu_a = 0.1\,\mathrm{mm}^{-1}$ and $\mu_s' = 0.4\,\mathrm{mm}^{-1}$. In view of the scattering phase function we consider the Henyey-Greenstein function with $g = 0.2$.

For the next comparison, we take into account the second-order moment (19) in the time domain. The absorption coefficient and the reduced scattering coefficient are, respectively, given by $\mu_a = 0.01\,\mathrm{mm}^{-1}$ and $\mu_s' = 0.9\,\mathrm{mm}^{-1}$. The anisotropy factor of the Henyey-Greenstein phase function is

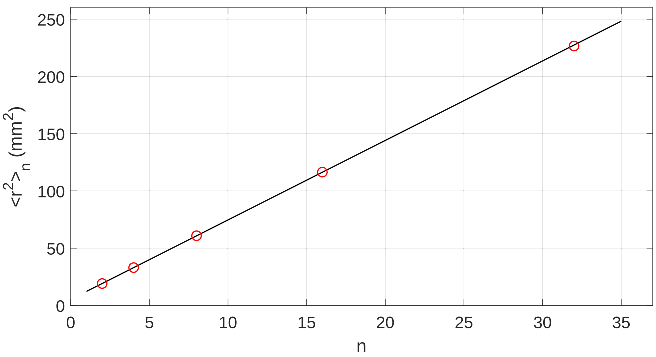

FIG. 1. MSD $\langle r^2 \rangle_n$ of the steady-state fluence $\Phi_n(\mathbf{x})$ as function of the scattering order.

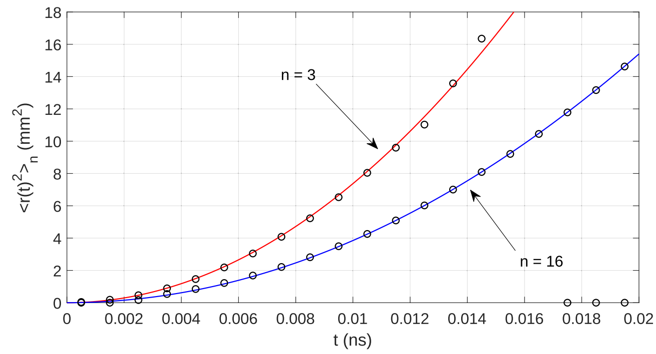

FIG. 3. MSD $\langle r^2(t) \rangle_n$ of the time-dependent fluence $\Phi_n(\mathbf{x}, t)$ for two different scattering orders.

set to $g = 0.8$. Figure 2 illustrates the analytical solution (solid lines) and the data predicted by the MC method (open circles) for two different scattering orders. Both solution approaches deliver within the statistical uncertainty of the MC method the same results.

Next, we use the same optical properties as for the last comparison to verify the time-dependent MSD (20). Figure 3 depicts the analytical solution (solid lines) and the MC data (open circles) for two different scattering orders. In accordance with the previous comparison, we obtain within the stochastic nature of the MC simulation an excellent agreement.

IV. DISCUSSION

In this article, we derived the exact second-order moment and the MSD for the transport theory fluence in the steady state and time domain as a function of the number of scattering events. In this context, we reduced the problem to a linear second-order difference equation that can be solved similarly to an ordinary differential equation. The resulting formulas, which are general in view of the anisotropic point source and the rotationally invariant scattering phase function, were compared with Monte Carlo simulations, showing within the stochastic nature of the Monte Carlo method a good agreement.

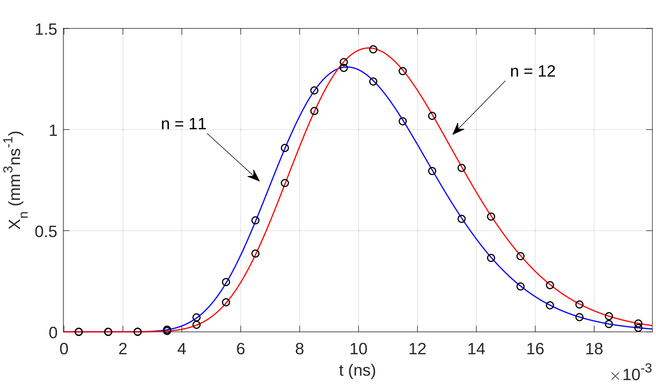

FIG. 2. Second-order moment $X_n(t)$ of the time-dependent fluence $\Phi_n(\mathbf{x}, t)$ for two different scattering orders.

The formulas for the MSD averaged over all scattering interactions were derived already in the literature, both in the steady state and time domain. However, as discussed above, the derivation was performed only for very restricted cases. Our derivation based on transport theory considering the general case of arbitrary rotationally invariant scattering phase functions including absorption shows that these equations have a much broader validity. Thus, we showed that the applicability of the well-known formulas reported by Einstein and Langevin are much larger compared to the restricted cases they were derived for. Further, the formula for the MSD versus the collision number also has important applications. For example, when light is propagating through random media which consist of moving particles, then, at each scattering event, the light frequency is shifted and, as a consequence, the total frequency shift depends on the number of interactions. The same argumentation holds true for Raman scattering. Furthermore, the MSD for fluorescence light depends on the interaction number, at which the fluorescence photon is generated. In view of the fluorescence, we additionally derived the second-order moment and the MSD in the steady-state domain. It should be noted that the steady-state formula for the MSD as a function of the number of scattering interactions was presented in the literature only for a restricted case, whereas the corresponding equation in the time domain could not be found in the literature. We additionally note that the MSD in the time domain depends not only on the product $\mu_s c$, but also on μ_s, see Eq. (21). Thus, if the time-resolved MSD is experimentally accessible, it is possible to derive both the effective scattering coefficient and the particle velocity in the considered medium applying the derived formula. Contrarily, in the often-used limiting value of this equation for long times it is only possible to derive the combination μ_s/c of both quantities, which is proportional to the diffusion coefficient. In addtion to the physical applications of the MSD, the derived expressions are also usable and important for the verification of MC methods [23].

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