Generalization of Fermat's principle for photons in random media: The least mean square curvature of paths and photon diffusion on the velocity sphere

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Photon migration in highly forward scattering random media can be described as a non-Euclidean diffusion (NED) on the velocity sphere. An exact path-integral solution of the corresponding NED equation in the photon five-dimensional phase space has been obtained. The solution leads to a ''generalized Fermat principle'' (GFP) for the most probable photon paths in turbid media: GFP requires the least mean-square curvature of the path. An explicitly analytic description of an ultrashort laser pulse propagation in random media based on NED equation is presented. Experiments have been performed to verify the NED theory. [S1063-651X(96)02205-2]

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The literature on the problem of electromagnetic wave propagation in random media is vast (see, e.g., [1]). The theory of this phenomena has been well elaborated especially for wave propagation in randomly inhomogeneous media in the small-angle scattering regime, when the characteristic scale of inhomogeneities $L \gg \lambda$, the wavelength; for recent and previous papers see Ref. [2]. These works address the case very important in space and atmosphere physics, when single and multiple scattering angles are always small and therefore an approximation for the wave equation, the so-called Leontovich stochastic parabolic equation (PE) [2], holds. A PE's counterpart, the small angle approximation (SAA) for the radiative-transfer equation (see, e.g., [1,3]), has the same restrictions. This theory is not applicable for the strong deflection angle regime, e.g., backscattering. Furthermore, both the PE and SAA theories are limited to the steady-state case. With the advent of ultrashort laser and enhanced ultrafast time-resolved registration techniques, new fundamental questions beyond the scope of these previously established techniques can be addressed. Among them is the problem of ultrashort laser pulse propagation in dense, highly forward-scattering media (important examples are bead suspensions, intralipid aqueous solutions, dense smog and fog, smoke, and biological tissues). The main feature of this problem that makes both the PE- and SAS-type approaches inapplicable is the strong deflection of photons from the direction of the incident pulse (sometimes called the ''diffusion'' regime). However, as far as photon migration on spatial scales less than $\sim 7 l_t$, the photon transport mean free path, is concerned, the conventional diffusion approximation was experimentally proven to fail for such media for the steady-state regime in Ref. [4] and for ultrashort laser pulses propagation in Ref. [5].

An important step beyond the conventional diffusion scheme was made in Ref. [6], in which a telegrapher's equation was introduced to optical physics. However, from Ref. [7] one concludes, that the telegrapher's equation is strictly valid in the region where the corrections to the diffusion equation are small. A further step to consider arbitrary multiple scattering angles in the frame of the radiative-transfer (Boltzmann) equation was undertaken in Ref. [8]. Still, a formal path-integral solution obtained in Ref. [8] was restricted to the scattering angle less than $\pi/2$. This path-integral solution is much too difficult to use for a practical analysis. Actually, it is expressed in terms of an unknown functional associated with the so-called pseudo-Fourier representation of the scattering phase function. Indeed, the final results of Ref. [8] upon absorption effects on radiative transfer are limited to the same region of applicability as the PE and the SAS. Recently, some attempts to attack again the concept of photon paths in turbid media have been undertaken. In Ref. [9] the Monte Carlo approach was used to simulate paths of photon migration. The backscattering for a highly forward scattering medium was addressed in Ref. [10]. Unfortunately, to avoid a cumbersome numerical solution of variational equations, these authors oversimplified the problem by using model expressions for the most probable photon paths, which violate the invariability of the speed of light in a homogeneous random medium.

In this paper, we introduce a non-Euclidean diffusion (NED) equation, which is a kind of general Boltzmann equation for photons in highly forward-scattered media with the collision integral represented as the non-Euclidean diffusion in the velocity space. The NED describes the important specific regimes of photon migration: the near ballistic and ''snake,'' developed diffusion regime and the transitional case corresponding to strong multiple scattering angles. An exact path-integral solution of the NED equation is presented, which particularly leads to the ''generalized Fermat principle'' (GFP) for the most probable photon paths in random media.

The conventional Fermat principle for transparent media selects the path between a source and a detector possessing the shortest photon optical paths length. The GFP for uniform random media selects among all photon paths possessing the same length, photon launching point and direction, as well as terminal point and direction, the most probable one characterized by the least mean-square curvature. An exact analytic solution for the ''Fermat'' paths and an approximate analytic intensity temporal profiles describing ultrashort laser pulse propagation in highly forward-scattering media are presented and briefly discussed. The NED theory has been verified by special time-resolved measurements. Experimental data favor the NED approach even in the region where diffusion theory was commonly believed to hold.

Photon migration in a macroscopically homogeneous random medium characterized by the speed of light c apparently can be described as a random walk on a spherical surface of the radius c, non-Euclidean velocity space S_2. For a medium composed of highly forward directed scatterers, the mean-square value of the deflection angle in a single collision event is given by $\langle\Delta^2\rangle\approx 2[1-\langle\cos(\Delta)\rangle]\ll 1$. After each collision a photon undergoes a small velocity change step $\delta s = c\sqrt{\langle\Delta^2\rangle}$ in a random direction on a small almost flat spherical segment of S_2. Given the photon collision frequency $1/\tau$, the photon random walk in the velocity space is simply diffusion with an effective diffusion coefficient $D_s=(\delta s)^2/4\tau=c^2\langle\Delta^2\rangle/4\tau=c^3/2l_t$, where l_t is the photon transport mean free path [note that $D_s=c^4(6D)^{-1}$, where $D=cl_t/3$ is the usual photon diffusion coefficient]. As a result, the photon distribution function $n(t,\vec{r},\vec{s},\vec{s}_0)$ satisfies the non-Euclidean diffusion equation in the photon phase space:

$$\frac{\partial n}{\partial t}+\vec{s}\vec{\nabla}_r n-\frac{c^4}{6D}\Delta_s n+\nu_a n=\delta(t)\,\delta(\vec{r})\,\delta(\vec{s}-\vec{s}_0), \qquad (1)$$

where ν_a is the absorption collision frequency and the right-hand side of Eq. (1) simulates an incident light pulse.

A proof is given below that the probability of each photon path [given by a photon velocity function $\vec{s}(t)$] is determined by $\kappa(t)$, the curvature of the photon path in real three-dimensional space, $c^2\kappa(t)=|d\vec{s}/dt|$, and that the solution of Eq. (1) for constant D and ν_a can be represented in the form of a path integral as

$$n(t,\vec{r},\vec{s},\vec{s}_0)=e^{-\nu_a t}\int\prod_\tau d\vec{s}(\tau)\,\delta\left(\vec{r}-\int_0^t\vec{s}(t')dt'\right)\times\exp\left(-\frac{3D}{2}\int\kappa^2(t)dt\right). \qquad (2)$$

One should not confuse this description with the completely different summation-over-paths picture of the conventional diffusion in the coordinate space. Note, for instance, that the exponential term in Eq. (2) contains the factor D, while the corresponding exponential term in the path-integral solution of the conventional diffusion equation [11] contains the factor D^{-1}.

A finite-difference representation of the function $n_1=n\exp(\nu_a t)$ resulting from Eq. (2) is

$$n_1(N,\vec{r},\vec{s},\vec{s}_0)=\frac{1}{A}\prod_{k=1}^{N-1}\left(\int_{S_2}\frac{d\vec{s}_k}{A}\right)\delta\left(\vec{r}-\varepsilon\sum_{k=1}^{N}\vec{s}_k\right)\times\exp\left(-\frac{\chi}{\varepsilon}\sum_{k=1}^{N}(\vec{s}_k-\vec{s}_{k-1})^2\right), \qquad (3)$$

where $t=N\varepsilon$, $A=\pi\varepsilon/\chi$, $\chi=l_t/2c^3$, $\vec{s}_k$ denotes the velocity at the time $t=k\varepsilon$, and $\vec{s}_N\equiv\vec{s}$. Each integration in Eq. (3) is performed over the spherical surface S_2 of the radius c. When one omits the δ function in Eq. (3) or (2), the function $n_1(t,\vec{r},\vec{s},\vec{s}_0)$ corresponds to a solution of the Schrödinger equation with the imaginary Planck constant for a particle moving on a sphere [11]: $(d\vec{s}/dt)^2$ stands for the kinetic energy of the particle.

Representing the δ function in Eq. (3) as $\delta(\vec{r}-\varepsilon\Sigma_{k=1}^{N-1}\vec{s}_k-\varepsilon\vec{s})$ and using the first-order Taylor expansion with respect to $\varepsilon\vec{s}$, one obtains a recurrent relation, which can be represented in the limit $N\gg 1$ as

$$n_1(N,\vec{r},\vec{s},\vec{s}_0)\approx\int_{S_2}\frac{d\vec{s}_{N-1}}{A}\exp\left(-\frac{\chi}{\varepsilon}(\vec{s}-\vec{s}_{N-1})^2\right)\times n_1(N-1,\vec{r},\vec{s}_{N-1},\vec{s}_0)-\varepsilon\vec{s}\,\vec{\nabla}_r\int_{S_2}\frac{d\vec{s}_{N-1}}{A}\times\exp\left(-\frac{\chi}{\varepsilon}(\vec{s}-\vec{s}_{N-1})^2\right)\times n_1(N-1,\vec{r},\vec{s}_{N-1},\vec{s}_0). \qquad (4)$$

In the limit $\varepsilon\to 0$ the second integral on the right-hand side of Eq. (4) reduces to $-\varepsilon\vec{s}\vec{\nabla}_r n_1(t,\vec{r},\vec{s},\vec{s}_0)$. Only the first integral term would appear in Eq. (4) if the Schrödinger equation for a particle on a sphere were considered. So one can handle the first integral in Eq. (4), recalling the above analogy with the Schrödinger equation. This means that the first integral in Eq. (4) reduces to $n_1(N-1,\vec{r},\vec{s},\vec{s}_0)+\varepsilon/(4\chi)\Delta_S n_1(N-1,\vec{r},\vec{s},\vec{s}_0)$, where Δ_S is the spherical part of the Laplace operator acting upon $\vec{s}$. The use of the apparent relation for the derivative $\partial n_1/\partial t=[n_1(N)-n_1(N-1)]/\varepsilon$ then would lead to the Schrödinger equation on a sphere if the second term on the right-hand side of Eq. (4) were neglected. When the latter is yet taken into account, one immediately arrives at the conclusion that the function $n=n_1\exp(-\nu_a t)$ satisfies Eq. (1).

According to Eq. (2), the probability of a particular path from the branch of all paths, connecting a collimated source and a detector and possessing the same path length ct, as well as initial $\vec{s}_0$ and terminal $\vec{s}$ propagation directions, is determined by its mean-square curvature $(1/t)\int_0^t\kappa^2(t')dt'$. Therefore, the most favorable path corresponds to the least mean-square curvature and satisfies the equation

$$\frac{\delta}{\delta\vec{s}(t)}\int_0^t dt'[(d/dt')^2-\lambda(t')\vec{s}^2(t')-\vec{C}\cdot\vec{s}(t')]=0. \qquad (5)$$

Here $\lambda(t)$ and $\vec{C}$ are the Lagrange multipliers corresponding, respectively, to the speed of light invariability condition $|\vec{s}(t)|=c$, and the arrival point restriction $\int_0^t\vec{s}(t')dt'=\vec{r}$. When the source and detector are chosen so that $\vec{s}_0,\vec{r},\vec{s}$ lie on the same plane, Eq. (5) can be solved exactly analytically. This leads to the following general expressions for the most probable paths given in a parametric form:

$$\Psi=\tau\int_{\gamma_0}^{\gamma}\frac{f_\Psi(\alpha-\varphi)d\alpha}{(\delta\sin\alpha\pm 1)^{1/2}}, \qquad (6)$$

where Ψ stands for x, y, or z, spatial coordinates or t, the time corresponding to a current point on the path, with α being a parameter on the path curve and $f_x(\alpha)=c\cos\alpha$, $f_y(\alpha)=c\sin\alpha$, $f_z(\alpha)=0$, and $f_t(\alpha)=1$. Arbitrary constants

$\tau,\delta,\varphi,\gamma_0$ are to be determined by the initial and final conditions. The details of the derivation of Eq. (6) have been discussed elsewhere [12]. The validity of Eq. (6), however, can be verified just by straightforwardly substituting it into Eq. (5).

There are few known examples of physical equations possessing an exact tractable path-integral solution with a clear and straightforward physical interpretation: the diffusion (heat transfer) equation and its quantum mechanical analog, the Schrödinger equation [11]; the telegrapher's equation and its quantum mechanical counterpart, the Dirac equation [13]; and an equation essentially very close to the diffusion equation, the Fokker-Planck equation [14]. The transport equation (1) adds a fundamentally and practically important member to this small family. The NED equation, Eq. (1), is apparently not a kind of Fokker-Planck equation since the third term on the left-hand side of Eq. (1) cannot be represented as a divergence. Despite that, Eq. (1) satisfies the law of conservation of particles as it should be. This follows from the fact that $\int_{S_2} d\vec{s}\ \Delta_s f(\vec{s}) \equiv 0$ for an arbitrary function f defined on a spherical surface S_2. Note that the most probable paths for particles moving according to the Fokker-Planck (or diffusion) equation with constant coefficients are infinite straight lines [14] in striking contrast to Eq. (6).

Earlier an approximate analytic description of short pulse propagation in random media based on a modified diffusion (telegrapher) equation was presented in Ref. [6], resulting in isotropic photon distribution. As a simple illustrative example of how the NED equation, Eq. (1), helps to describe anisotropy effects, we now present an approximate analytic solution of Eq. (1) for the function representing photon number density

$$N(t,\vec{r},\vec{s}_0) = \frac{1}{c^2}\int_{S_2} d\vec{s}\ n(t,\vec{r},\vec{s},\vec{s}_0)$$
$$= \frac{1}{(2\pi)^3 c^2}\int_{S_2} d\vec{s}\int d\vec{k}\left\langle \exp\left(i\vec{k}\int_0^t \vec{s}(t)dt - i\vec{k}\vec{r}\right)\right\rangle, \tag{7}$$

expressed in terms of the random vector function $\vec{s}(t)$ with the averaging defined in terms of a path integral by Eq. (2).

Equation (7) can be evaluated using standard cumulant decomposition interrupted after the second term. The calculations are lengthy but straightforward and will be presented elsewhere. We thus obtain

$$N(t,\vec{r},\vec{s}_0) = \frac{1}{(4\pi)^{3/2}}\frac{1}{\sqrt{\det B}}\exp(-\tfrac{1}{4}B^{-1}_{\alpha\beta}(r-a)_\alpha (r-a)_\beta),$$

$$\vec{a} = \frac{\vec{s}_0 l_t}{c}(1-e^{-\tau}),$$

$$B_{\alpha\beta} = \frac{l_t^2}{2}\delta_{\alpha\beta}[\tfrac{2}{3}\tau - (1-e^{-\tau}) + \tfrac{1}{9}(1-e^{-3\tau})]$$
$$+ \frac{l_t^2}{2c^2}(s_0)_\alpha (s_0)_\beta[(1-e^{-\tau}) - \tfrac{1}{3}(1-e^{-3\tau})$$
$$-(1-e^{-\tau})^2], \tag{8}$$

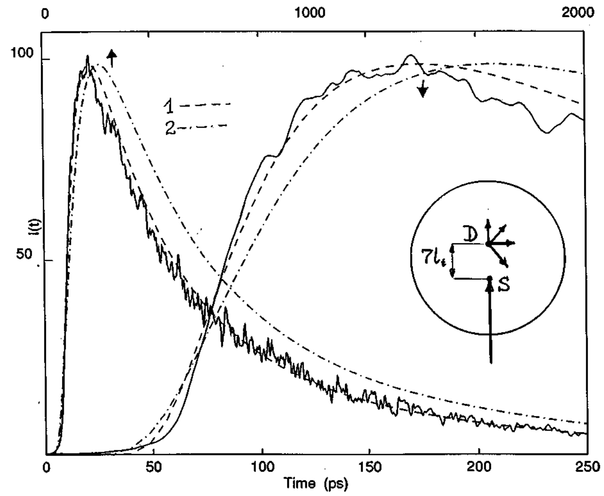

FIG. 1. Normalized intensity temporal profiles $I(t) = I(t,r,\vec{s}_0)$ or $N(t,\vec{r},\vec{s}_0)$, measured and calculated using the NED and the conventional diffusion approximation for the source-detector distance $7l_t$ (see the text for more details). Wavy solid lines, the experimental results; curve 1, Eq. (8); curve 2, Green's function for the conventional diffusion equation. The rise part and entire pulse are depicted separately (arrows indicate the corresponding time axes). The inset shows the experimental geometry. Source (S) is an optical fiber with a divergence angle of 5°. The orientation of the receiver (D) (a fiber with 15° collection angle) was changed to detect scattered light propagating in different directions.

where $\tau = ct/l_t$ is the reduced time and B^{-1} denotes the matrix reciprocal to B.

The following interpretation of Eq. (8) can be given. It describes the evolution of a photon "cloud" initially concentrated at the origin and launched in the direction $\vec{s}_0$. The center-of-mass motion of the cloud is described by the function $\vec{a}(t)$. The spreading of the cloud is described by the matrix $B(t)$. For small times $t \ll 1$ one has $\vec{a}(t) = \vec{s}_0 t$, which corresponds to the near ballistic photon motion. According to Eq. (8) in the near ballistic regime, the spreading of the photon "clot" in the $\vec{s}_0$ (longitudinal) direction is governed by the law $\Delta x_l \sim t^2$ and in the transversal direction $\Delta x_{\rm tr} \sim t^{3/2}$. These are considerably slower than ordinary diffusion law $\Delta x_d \sim t^{1/2}$. For large times $\tau \gg 1$ Eq. (8) describes the ordinary diffusion spreading with respect to an imaginary point source shifted from the origin by the vector $\vec{a} = \vec{s}_0 l_t / c$.

To support the NED theory at spatial scales where the diffusion approximation is commonly supposed to hold, the experiment was performed using a 100-fs laser pulse at 625 nm, a streak camera detection system with ~10-ps time resolution, and 0.132 vol % aqueous suspension of polystyrene spheres (0.303 ± 0.0057 μm in diameter, $g = \langle \cos\Delta \rangle \approx 0.7$), contained in a 10-cm cylindrical tank. For such a concentration l_t was experimentally shown to be exactly proportional to the concentration of scatterers [15]. The calculations lead to the Mie-based value of $l_t = 2.00 \pm 0.04$ mm. The effective absorption length $l_a = c/\nu_a$ was estimated to be 700 ± 150 mm. Time-resolved intensities $I(t,\vec{r},\vec{m},\vec{s}_0)$ were measured in the photon launching direction at $7l_t$ from the source for different orientations $\vec{m}$ of detecting fibers (Fig. 1). The intensity $I(t,\vec{r},\vec{m},\vec{s}_0)$ can be related to the photon distribution function $n(t,\vec{r},\vec{s},\vec{s}_0)$ as $I(t,\vec{r},\vec{m},\vec{s}_0)$

$=\int A(\vec{m},\vec{s})n(t,\vec{r},\vec{s},\vec{s}_0)d\vec{s}$, where $A(\vec{m},\vec{s})$ is the so-called receiving cross section [1]. The corresponding experimental data $I(t,\vec{r},\vec{m},\vec{s}_0)$ were combined to obtain the angle-averaged profile $I(t,\vec{r},\vec{s}_0)=\int d\vec{m}I(t,\vec{r},\vec{m},\vec{s}_0)=\widetilde{A}N(t,\vec{r},\vec{s}_0)$, where $\widetilde{A}=\int A(\vec{m},\vec{s})d\vec{m}$ is a constant characterizing the fiber. This condition makes possible the comparison of Eq. (8) with the experimental data. A particular distance $7l_t$ was selected for the experiment since the conventional diffusion theory is commonly supposed to be valid at such distances and it was demonstrated to hold within 1% accuracy for the net transmission through a slab thicker than $5l_t$[16]. The experimental intensity temporal profile $I(t,\vec{r},\vec{s}_0)$ is shown in Fig. 1 along with two theoretical curves, based on Eq. (8) and the conventional diffusion model, calculated using the Mie-based $l_t=2.00$ mm and $l_a=700$ mm. The rise part and the peak region of the temporal profile are not sensitive to l_a within the given range. It is seen that experimental data definitely favor the NED theory in spite of the large source-detector distance used in measurements and g not very close to 1.

The GFP approach may have a potential to simplify the solution of the object recognition problem for optical tomography. Different photon paths between a source S and a detector D (both well collimated to emit and collect light in a narrow solid angle) corresponding to a detection time slice $T,T+dT$ have the same lengths and tangents at the S and D points, but the Fermat path, Eq. (6), has the largest probability. If an obstacle crossed this particular Fermat path, it would lead to the largest change in the detector readings. By varying the source and detector positions the whole medium can be covered by a grid formed as a set of intersecting Fermat paths (like coordinate lines on a map). Placed somewhere inside the medium, an absorbing foreign object would cover certain grid nodes and its location can be determined as a crossing points of the corresponding Fermat paths from comparative intensity measurements. Though this imaging scheme, introducing the idea of utilizing collimated sources and detectors and the correspondence between Fermat photons paths and source-detector configurations, looks simple and attractive, one important question is the accuracy of resolution, which depends on the effective width W_r of the branch of paths concentrated around a Fermat path. Since a collimated detector selects a certain group of paths, while terminating the others arriving at the same location and time but at different angles, one can expect W_r to be smaller than the diffusion regime would assume.

The non-Euclidean diffusion equation in the photon velocity space can be important for many challenging applications of ultrafast optical phenomena, when the spatial resolution of the order of l_t is of interest such as, for instance, in optical imaging ($l_t\sim 1$ mm for biological tissues). We also believe that the nontrivial feasibility to observe well defined curvilinear photon paths in multiply scattering media and to interpret the respective intensity temporal profiles will attract the attention of groups working at the frontier of ultrafast laser optics of random media for optical tomography.

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[1] A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic, New York, 1978).

[2] V. I. Tatarskii, A. Ishimaru, and V. U. Zavorotny, *Wave Propagation in Random Media (Scintillation)* (SPIE, Washington,1993); V. I. Tatarskii and V. U. Zavorotnyi, Prog. Opt. **18**, 205 (1980); R. Dashen, J. Math. Phys. **20**, 894 (1979); V. I. Tatarskii, *The Effect of Turbulent Atmosphere on Wave Propagation* (National Technical Information Service, Springfield, VA, 1971).

[3] L. Dolin, Radiophys. Quantum Electron. **9**, 40 (1973); R. L. Fante, IEEE J. Quantum Electron. **AP-21**, 750 (1973).

[4] A. Ishimaru, Y. Kuga, R. L.-T. Cheung, and K. Shimizu, J. Opt. Soc. Am. **73**, 131 (1983).

[5] K. M. Yoo, F. Liu, and R. R. Alfano, Phys. Rev. Lett. **64**, 2647 (1990); **65**, 2210(E) (1990).

[6] A. Ishimaru, J. Opt. Soc. Am. **68**, 1045 (1978); Appl. Opt. **28**, 2210 (1989).

[7] K. Furutsu, J. Opt. Soc. Am. **70**, 360 (1980).

[8] J. Tassendorf, Phys. Rev. A **35**, 872 (1987).

[9] Sh. Feng, F. Zeng, and B. Chance, SPIE Proc. **1888**, 78 (1993).

[10] L. T. Perelman, J. Wu, I. Itzkan, and M. S. Feld, Phys. Rev. Lett. **72**, 1341 (1994).

[11] R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965); Chap. 12; M. Kac, *Probability and Related Topics in Physical Science* (Interscience, London, 1957).

[12] A. Ya. Polishchuk and R. R. Alfano, Opt. Lett. **20**, 1937 (1995).

[13] C. DeWitt-Morette and S. K. Foong, Phys. Rev. Lett. **62**, 2201 (1989); B. Gavean, T. Jacobson, M. Kac, and L. S. Shulman, *ibid.* **53**, 419 (1984).

[14] H. Haken, Z. Phys. B **24**, 321 (1976).

[15] G. H. Watson, S. L. McCall, P. A. Fleury, and K. B. Lyons, Phys. Rev. B **41**, 10 947 (1990).

[16] D. J. Durian, Phys. Rev. E **50**, 857 (1994).