Numerical predictions of short-pulsed laser transport in absorbing and scattering media. II – A frequency-based approach.

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The one-dimensional transient radiative transfer problem consider in [1] is computationally solved here by the use of a Finite Volume method formulated in the space-frequency domain instead of the space-time domain . It has been shown in [1] that numerical approaches based in the space-time domain induce transmitted flux emerging earlier than the minimal time required by the radiation to leave the medium. Therefore, to eliminate the time dependence, a frequency-based numerical method is formulated, implemented, and validated in this paper. Results for transmittances are accurate, without physically unrealistic behaviors at early time periods. However, the frequency-dependent approach is computationally relatively expensive: it requires approximately five times more computational time than its temporal counterpart.

Keywords: radiative transfer; transient regime; short laser pulse; frequency-domain approach; finite volume method

NOMENCLATURE

a asymmetry factor of the linear scattering phase function, -

c speed of electromagnetic wave, ms^{-1}

H Heaviside function, -

i complex constant, $(-1)^{1/2}$

I intensity, $Wm^{-2}sr^{-1}\mu m^{-1}$

j spatial node, -

k frequency step, -

m discrete direction, -

n temporal step, -

Re real part of a complex number, -

S_c source function due to the scattering of the collimated component of intensity, $Wm^{-2}\mu m^{-1}$

t time, s

t^* dimensionless time, -

T hemispherical transmittance, -

w_m angular weight, solid angle, -

Greek letters

β extinction coefficient, m^{-1}

θ polar angle, rad

μ	direction cosine relative to the polar angle, -
τ	optical depth, -
τ_L	optical thickness, -
Φ	scattering phase function, -
ω	albedo for single scattering, -
$\hat{\omega}$	time-dimensionless angular frequency (pulsation), rad

Subscripts/Superscripts

$'$	other directions
^	frequency-dependent quantity
0	incident collimated beam
c	collimated component
d	diffuse component
j	refers to a spatial node
$j \pm ½$	refers to the boundaries of a control volume
k	refers to a discrete frequency step
L	refers to medium thickness
m	refers to a discrete direction
n	refers to a temporal step
p	refers to the pulse

1. INTRODUCTION

In the previous paper [1], the one-dimensional transient radiative transfer problem for absorbing and scattering media has been solved in the space-time domain using a Finite Volume Method (FVM). The Van Leer and Superbee flux limiters, based on the second order spatial scheme Lax-Wendroff, have been used in order to minimize the transmitted fluxes emerging before the minimal time required by the radiation to leave the medium. Results for transmittance have shown that these non-physical results cannot be completely avoided with time-based methods, due to the interdependence between the spatial and temporal discretizations.

In order to assess the use of the Diffusion approximation for the solution of transient radiation transport problems, Elaloufi et al. [2] solved the time-dependent radiative transfer equation in the space-frequency domain with a DO method. Therefore, the main objective of this paper is to embed such an idea within the FVM and test it.

Boulanger and Charette [3,4] have recently proposed transient inversion algorithms in order to map the optical properties of an absorbing and scattering medium irradiated by a short pulsed near infra-red laser, given a set of measurements at the boundaries. The authors pointed out that this indirect optical spectroscopy technique, or tomography, is highly dependent on the model used to track radiative energy propagation in participating media, which motivated the work done in this paper.

The theoretical FVM formulation is summarized in the second section: the assumptions are recalled for convenience, the Complex Radiative Transfer Equation (CRTE) is derived, and the FVM formulation in the frequency-domain is discussed. The method is then applied to two typical 1D transient radiative transfer problems, and results are compared with those obtained in the companion paper [1] and from those available in the literature. Finally, the application of the CRTE to a frequency-based optical diagnostic technique is discussed.

2. THEORETICAL AND NUMERICAL FORMULATIONS

2.1. Transient radiative transfer problem description and assumptions

The problem description and related basic assumptions are readily available in [1].

2.2. Mathematical modeling in the space-frequency domain

In this work, the Transient Radiative Transfer Equation (TRTE) still describes spatial and temporal variations of the diffuse component of the intensity along the direction μ in an absorbing and scattering medium and has been derived in [1] using dimensionless time ($t^* = \beta ct$) and optical depth ($\tau = \beta z$) as variables.

Here, since the objective is to solve the transient radiative transfer problem in the space-frequency domain, all temporal variables have to be transformed into frequency-dependent variables by applying a temporal Fourier Transform (FT) of the intensity [2]:

$$I(\tau,\mu,t^*) = \int_{-\infty}^{\infty} \hat{I}(\tau,\mu,\hat{\omega}) \exp(i\hat{\omega}t^*) d\hat{\omega} . \tag{1}$$

where $\hat{\omega}$ is the angular frequency deriving from the dimensionless time variable t^*. Inversely the frequency-dependent intensity can be written as the FT of the time-dependent intensity. The time-dependent Fourier analysis applied to the TRTE leads to:

$$\mu \frac{d\hat{I}_d(\tau,\mu,\hat{\omega})}{d\tau} = -(1+i\hat{\omega})\hat{I}_d(\tau,\mu,\hat{\omega}) + \frac{\omega}{2}\int_{-1}^{1} \hat{I}_d(\tau,\mu',\hat{\omega})\Phi(\mu',\mu)d\mu' + \hat{S}_c(\tau,\mu,\hat{\omega}) . \tag{2}$$

where the frequency-dependent intensity $\hat{I}_d(\tau,\mu,\hat{\omega})$ and $(1+i\hat{\omega})$ are complex numbers. The first and second terms of the right-hand side of equation (2) represent respectively the attenuation by absorption and scattering and the reinforcement due to the scattering of the diffuse part of intensity. This equation has the form of a steady-state radiative transfer equation and is called the Complex Radiative Transfer Equation (CRTE). The source term $\hat{S}_c$ originating from the scattering of the collimated intensity is determined from a particular solution of equation (2) without scattering sources [2]:

$$\hat{S}_c(\tau,\mu,\hat{\omega}) = \frac{\omega}{4\pi}\hat{I}_0(\hat{\omega}) \exp[-\tau(1+i\hat{\omega})]\Phi(1,\mu) . \tag{3}$$

where $\hat{I}_0(\hat{\omega})$ is obtained from the temporal Fourier analysis of the incident pulse; this is schematically depicted in figure 1.

[Insert figure 1 about here]

From the above statement and figure 1, it is concluded that the solution of a transient radiative transfer problem can be obtained by solving the CRTE for each angular frequency contained in the temporal Fourier decomposition of the pulse.

2.3. Numerical solution in the space-frequency-domain

As for the time-dependent numerical solution presented in part [1], a FVM is used in order to solve the transient radiative transfer problem in the space-frequency domain. For a given angular frequency $\hat{\omega}$, a steady-state like radiation transport problem is solved by calculating the frequency-dependent intensities on the J nodes and M directions of the spatial and directional discretizations, respectively. Then, integrating the CRTE over a control space $\Delta\tau_j w_m$ yields:

$$\hat{I}_j^{m,k} = \frac{\dfrac{-\mu_m}{\Delta\tau_j}\left(\hat{I}_{j+1/2}^{m,k} - \hat{I}_{j-1/2}^{m,k}\right) + \dfrac{\omega_j}{2}\sum_{m'=1}^{M} w_{m'}\hat{I}_j^{m',k}\Phi_j^{m'm} + \hat{S}_{c_j}^{m,k}}{1 + i\hat{\omega}_k}. \qquad (4)$$

In the above, $\hat{I}_j^{m,k}$ is the frequency-dependent intensity at node *j*, in direction *m*, and at frequency *k*, corresponding to the discrete pulsation $\hat{\omega}_k$ for which the CRTE is solved. The subscript *d*, referring to the diffuse component of intensity, is omitted for more clarity. The spatial and directional solutions of the CRTE are exactly the same than for the TRTE [1]. For the frequency-dependent approach, a first order upwind scheme is sufficient for proper interpolation, since the CRTE is a steady-state equation.

Equation (4) is then solved, for a specific frequency *k*, for each node *j*, and in each direction *m* of the spatial and angular discretizations, respectively. An iterative scheme, based upon the convergence of the source term due to the scattering of the diffuse part of intensity, is used.

In order to determine the frequencies for which the CRTE is solved, a temporal Fourier Transform (FT) is applied on the incident square pulse:

$$I_0^k = I_0\left(\frac{\sin\hat{\omega}_k t_p^*}{\hat{\omega}_k} - i\frac{1-\cos\hat{\omega}_k t_p^*}{\hat{\omega}_k}\right). \tag{5}$$

The *K* relevant angular frequencies are selected by calculating equation (5) for a series of discrete pulsations $\hat{\omega}_k$. A graphical representation of the real and imaginary parts of I_0^k, as a function of $\hat{\omega}_k$, allows one to find the threshold angular frequencies, $\hat{\omega}_{-K/2}$ and $\hat{\omega}_{K/2}$, for which the CRTE has to be solved (i.e., frequencies below and above which the real and imaginary parts of I_0^k vanish). The angular frequency discretization $\Delta\hat{\omega}_k$ is determined, as for the spatial discretization, by performing a numerical sensitivity analysis [17].

Finally, the time-dependent intensities have to be recovered from the frequency-dependent intensities. This is done by applying an Inverse Fourier Transform (IFT) on the frequency-dependent intensities:

$$I_j^{m,n} = \mathrm{Re}\left\{2\sum_{k=0}^{K/2}\frac{1}{K}\exp(i\hat{\omega}_k t_n^*)\hat{I}_j^{m,k}\right\}. \tag{6}$$

Equation (6) implies that the frequency-dependent intensities can be calculated and summed only for the $K/2$ relevant angular frequencies (symmetry of the summation) in order to determine the time-dependent intensity at a given instant t_n^*. Unlike the space-time approach, the determination of the intensity at instant t_n^* doesn't require the knowledge of intensities at 0, t_1^*, t_2^*, …, t_{n-1}^*.

The principal steps to solve the transient radiative transfer problem in the space-frequency domain are summarized hereafter: (1) the temporal FT of the pulse is calculated; (2) the relevant frequencies are determined; (3) a CRTE is solved for each selected angular frequency; (4) an IFT is applied in order to derive time-dependent intensities.

3. NUMERICAL RESULTS

The two transient radiative transfer problems treated in part A are solved here with the frequency-dependent approach, and are recalled for convenience; in both cases, the absorbing and scattering medium is illuminated by a square pulsed collimated radiation beam of unit intensity ($I_0 = 1$) and unit dimensionless duration ($t_p^* = 1$) on its boundary at $\tau = 0$. When the medium is anisotropically scattering, the linear anisotropic phase function is considered ($\Phi(\mu,\mu') = 1 + a\mu\mu'$). In post-treatment, the temporal hemispherical transmittance is calculated from the time-dependent intensities, and can be written as:

$$T^n = \frac{2\pi \sum_{\mu_m > 0} w_m \mu_m I_J^{m,n} + I_{c_J}^{m_0,n}}{I_0}. \qquad (7)$$

The numerical implementations of the proposed formulation are written in *FORTRAN* and compiled with the software *Microsoft Developer Studio – Fortran PowerStation 4* on a single processor desktop PC.

3.1. Problem 1: Optically thick and highly scattering medium

This first problem is akin to that presented en [1]: the optical thickness of the medium is $\tau_L = 10$ while the scattering albedo is $\omega = 0.998$.

Also, the temporal Fourier transform of the square pulse allows the determination of pulsations for which the CRTE has to be solved (from -16π to 16π, except $\hat{\omega}_k$ = 0). A preliminary parametric analysis permitted the identification of an optimal angular frequency discretization ($\Delta\hat{\omega}_k = 3 \times 10^{-4}\pi$).

Transmittances obtained by solving the CRTE with a FVM and a first order upwind scheme are reported in figure 2. It is important to note that since the optical thickness (τ_L) of the slab is 10, the minimal dimensionless time required by the radiation to leave the medium (t_L^*) is also 10.

[Insert figure 2 about here]

Figure 2 clearly indicates that for a large time scale (from 0 to 100), results from the frequency-dependent method are in good agreement with those obtained from a reference Monte Carlo (MC) formulation [6], even at early time periods (i.e., near t_L^*). It is important to note here that the physical interpretation of the results is available in [1].

In order to analyze more precisely the relative accuracy of the frequency-dependent approach, the transmittances obtained from this method are compared at early time periods with those obtained with a first order exponential scheme [7], the Van Leer flux limiter and the Superbee flux limiter. Only results for a = 0.9 are shown, since early transmitted radiation becomes more important within a highly forward scattering medium.

[Insert figure 3 about here]

Figure 3 shows that no energy is transmitted before $t_L^* = 10$ with the frequency-dependent method : there is no interdependence between the spatial and temporal discretizations.

It can be concluded that, from a strict point of view of accuracy, the frequency-dependent method is superior than solving directly the TRTE in the space-time domain, since the transmittance begins exactly at the dimensionless time $t^* = 10$. Even after this instant, the approaches give different results, and converged to the same values approximately between $t^* = 10$ and $t^* \approx 12$. However, the temporal solution of the available MC results [6] is insufficient to conclude about this divergence.

The CPU times associated with the different methods are presented in table 1, and have been compiled for the case of an isotropic scattering medium ($a = 0$).

[Insert table 1 about here]

The frequency-dependent approach is time consuming : a large number of frequencies is required to model a square pulse, and the shift from the space-frequency to the space-time domain (IFT) constitutes a supplementary computational step, compared to the direct solution of the TRTE. For this particular problem, the frequency-dependent method requires approximately five times more CPU time than solving the problem in the space-time domain with a Superbee flux limiter.

3.2. Problem 2: Medium of unit optical thickness and variable scattering albedo

In this second problem, the optical thickness of the medium is fixed at $\tau_L = 1$ while the scattering albedo ω is variable (0.25, 0.50, 0.75 and 0.90). In all cases, the medium is isotropically scattering (a = 0). This second test problem has been solved with a Discrete Ordinates approach coupled with the Piecewise Parabolic Advection scheme (DO-PPA) [8], which has been validated with a Monte Carlo formulation [6]. The same spatial, angular, and temporal discretizations used for the first problem are found sufficient.

Transmittances from the frequency-dependent approach are presented in figure 4.

[Insert figure 4 about here]

As for the first problem, results from the space-frequency method are in good agreement with those from the literature [8]. It can also be seen that the physics of the problem is respected, since the transmittance begins exactly at t^* = 1. Notice that the physical significance of the above results is discussed in [1].

A comparison between the frequency and time-dependent methods (exponential scheme [7], Van Leer flux limiter, and Superbee flux limiter), at early time periods, is shown in figure 5.

[Insert figure 5 about here]

On contrary to what has been observed for the first problem (figure 3), figure 5 does not report significant differences between the two approaches after t_L^*. This can be explain by the fact that, for a medium of unit optical thickness, the ballistic regime (collimated component) is dominant compared to the sinuous propagation regime (diffuse component), in the beginning of the temporal process. Hence, the collimated intensity does not induced early transmitted radiation, since its solution is determined exactly by a spatial exponential decay. It can be seen in figure 4 that for dimensionless time t^* from 1 to 2 (temporal interval for which the pulsed collimated radiation beam is transmitted at $\tau = \tau_L$), the sinuous propagation regime has a low influence on the overall transmittance.

The CPU times associated with the different solution approaches are presented in table 2, when the scattering albedo is 0.9. Previous conclusions hold.

[Insert table 2 about here]

The CRTE has been applied here to solve transient radiative transfer problems. This mathematical model can also be used for an alternative frequency-based optical tomography technique; this is briefly overviewed in the next section.

4. RADIATIVE METROLOGY BASED ON THE CRTE

Optical tomography techniques refer to the use of low-energy visible or near infra-red light to irradiate highly scattering media in order to map their radiative properties, given a set of measurements at the boundaries (transmitted and/or reflected fluxes) [9]. To achieve that, the data can either be acquired as time-varying intensities (response to an ultra-short pulsed laser) as discussed in this paper, or by steady-state measurements (response to a continuous radiation beam).

Biomedical works have reported that traditional steady-state reconstruction algorithms suffer from considerable cross-talk between absorption and scattering coefficients [10,11]; this means that purely scattering (or absorbing) inclusions are reconstructed with unphysical absorption (or scattering) properties [12]. This cross-talk is caused by the fact that different distributions of radiative properties inside the medium can lead to the same value collected at the boundaries [12].

The time-dependent optical tomography techniques can prevent the cross-talk between radiative properties, since it allows the collection of several temporal data per measurement (unlike stationary techniques in which the measurement is limited to only one steady-state value of intensity). In counterpart, the main disadvantages of this approach are the small time scales to handle (temporal responses extend only over few nanoseconds), the high cost of the experimental devices [13], and their calibration problems [14].

Therefore, works originating from the biomedical area [8,12-15] suggest an alternative metrology based in the frequency-domain, in which the medium is illuminated by a continuous light source whose amplitude is modulated at one given frequency (typically at 100-1000 MHz [11,12]). A major advantage of this technique, unlike traditional steady-state approach, is that additional information is obtained by measuring both the amplitude (A) and the phase shift (φ). It is then expected that frequency-domain devices will permit a better separation of absorption and scattering coefficients at cheaper cost and efforts than time-dependent techniques [12]. These three different experimental techniques are schematically depicted in figure 6.

[Insert figure 6 about here]

The application of the frequency-domain optical tomography has been, since recently [12], restricted to the determination of radiative properties of optically thick and highly scattering media since the modeling has been often limited to the use of the Diffusion approximation. The CRTE presented in this paper, and reported in parallel by two other groups [11,16], allows the application of the space-frequency method to media that are not necessarily optically thick.

Finally, as schematically depicted in figure 1, a pulse can be treated as a superposition of continuous radiation beams for which the amplitude is modulated at one given frequency. It is clear from this statement that for a single source-detector pair, the time-dependent approach carries more information since a pulse contains many frequencies. However, Yodh and Chance [13] suggested that frequency-based optical diagnostic techniques that employ well-chosen modulation frequencies are technically simpler, while the same essential information is provided about the medium properties. Therefore, a preliminary sensitivity analysis has been performed by the authors to derive some optimal parameters for the design of a frequency-domain optical diagnostic technique based on the CRTE [17].

5. CONCLUSION

The one-dimensional transient radiative transfer problem for absorbing and scattering media has been solved, in Cartesian coordinates, in the frequency-dependent method using a Finite Volume Method.

It has been shown that the temporal distributions of transmittance obtained from the frequency-domain approach are accurate, without physically unrealistic fluxes emerging earlier than the minimal time required by the radiation to leave the medium. Due to the interdependence between the spatial and temporal discretizations, this precision can not be achieved with a space-time method, even if high order interpolation scheme and flux limiters are used, as shown is part A of this work.

However, compared to a time-dependent approach, the space-frequency method is time consuming (requires approximately five times more CPU time than solving directly the TRTE with a Superbee flux limiter). A Fast Fourier Transform (FFT) algorithm was implemented to accelerate the Inverse Fourier Transform (IFT) step, but the success of this strategy was mitigated: no significant improvement in CPU time was reported (unpublished). The reason is that the IFT itself is not what burdens the algorithms, and the large CPU time are mostly due to the large numbers of angular frequencies needed to correctly represent the square pulse. Therefore, a more physically realistic Gaussian incident pulse must be taken into account, since less angular frequencies will be required to adequately represent this temporal pulse.

Finally, the advantages of performing frequency-domain measurements over traditional steady-state and time-dependent devices have been pointed out. Therefore, research works are currently devoted to the development of inversion algorithms based on the Complex Radiative Transfer Equation for the design of such frequency-based optical diagnostic technologies.

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FIGURES AND TABLES

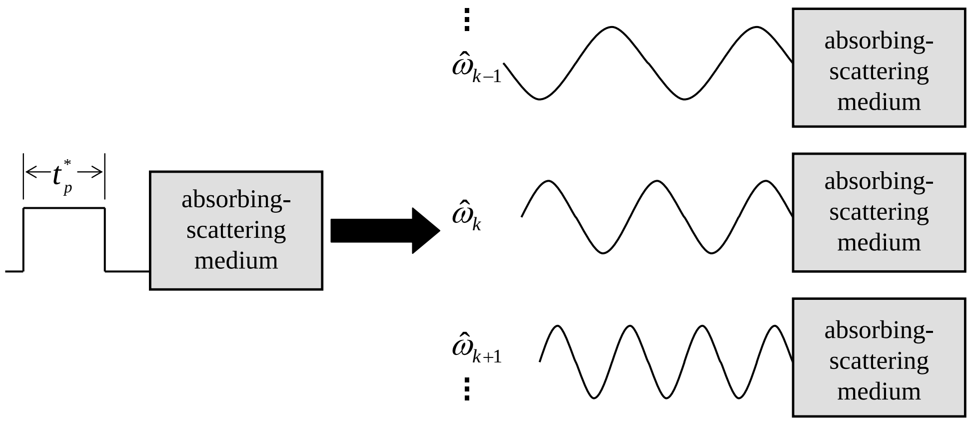

Figure 1: Shift from the space-time to the space-frequency domain.

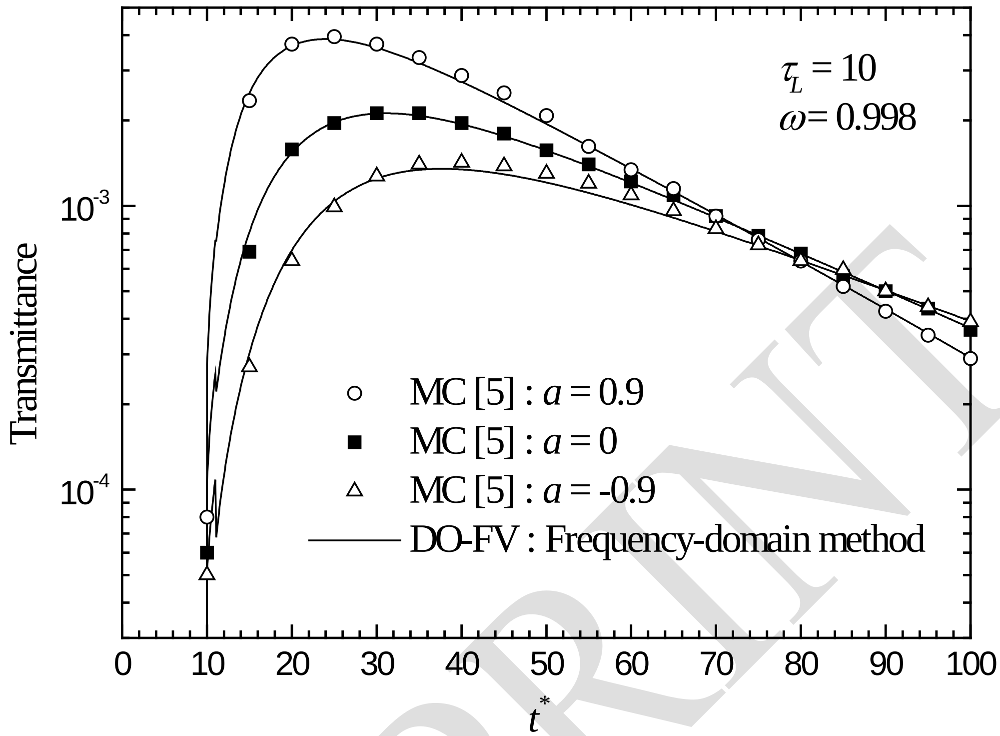

Figure 2: Temporal distribution of the hemispherical transmittance; comparison between the frequency-domain approach and a Monte Carlo formulation [5].

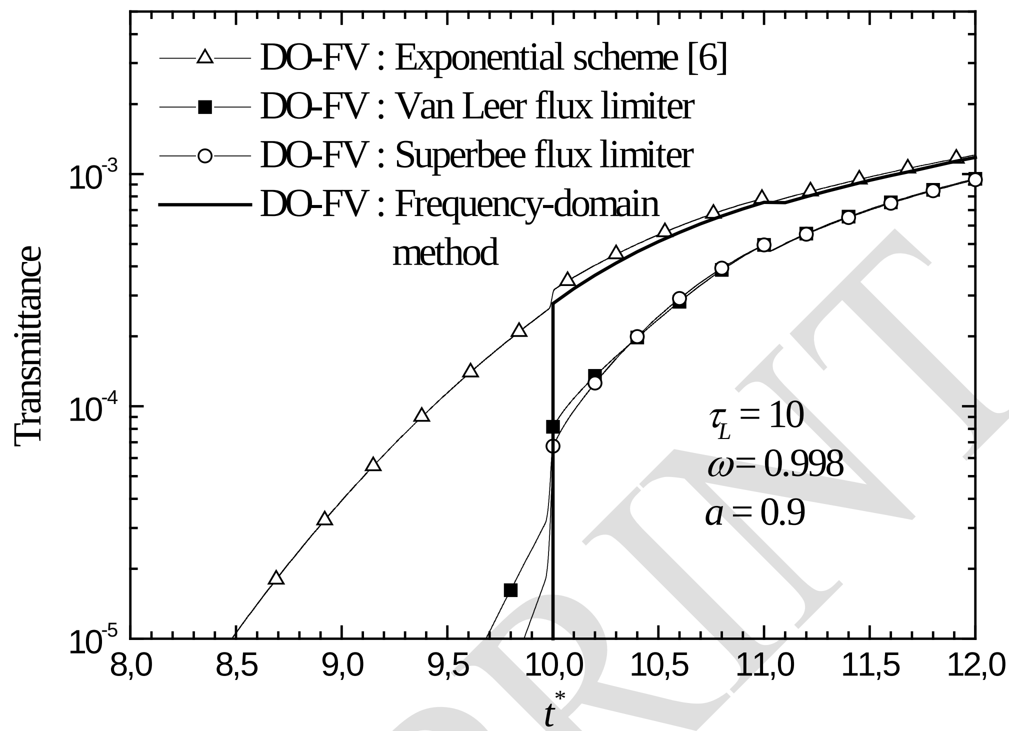

Figure 3: Temporal distribution of hemispherical transmittance; comparison between the Van Leer flux limiter, the Superbee flux limiter, the exponential interpolation scheme [6], and the frequency-domain method at early time periods.

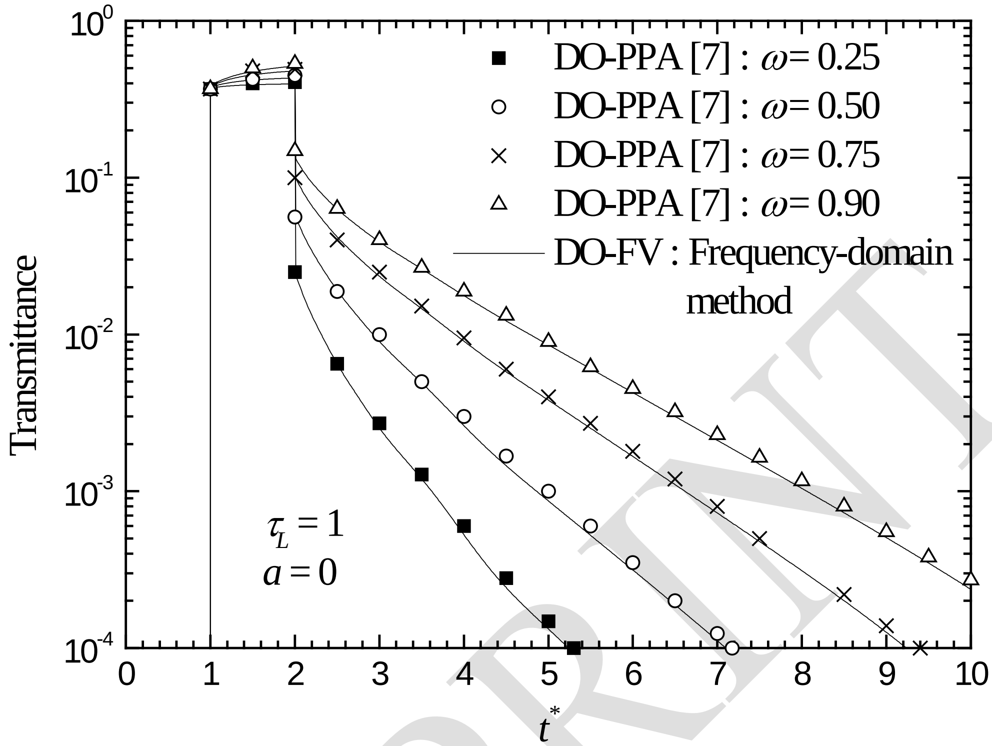

Figure 4: Temporal distribution of hemispherical transmittance; comparison between the frequency-domain approach and the DO-PPA method [7].

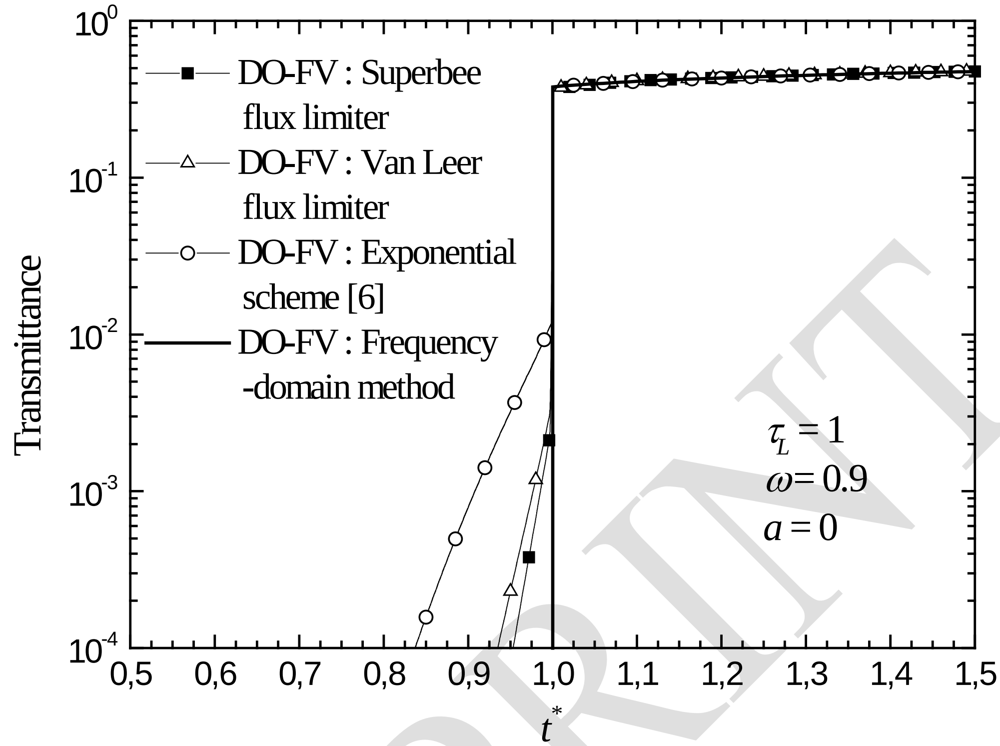

Figure 5: Temporal distribution of hemispherical transmittance; comparison between the Van Leer flux limiter, the Superbee flux limiter, the exponential interpolation scheme [6], and the frequency-domain method at early time periods.

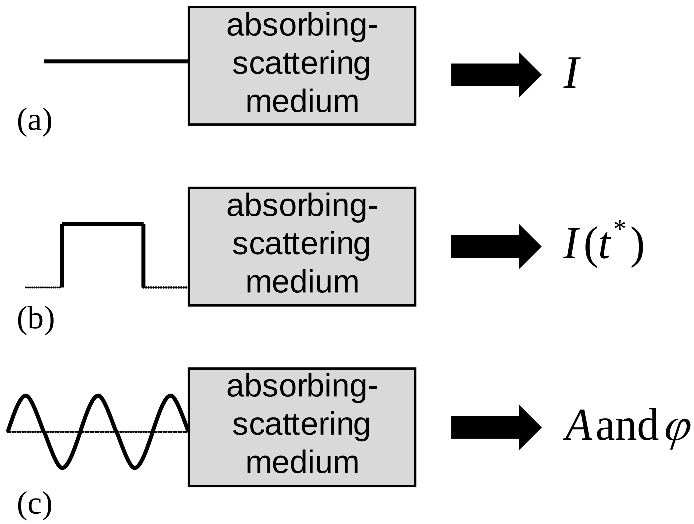

Figure 6: Schematic representation of optical tomography techniques: (a) traditional steady-state; (b) time-dependent; (c) frequency-based.

Resolution methods	CPU time [s]	Relative CPU time (Superbee flux limiter) [-]
Time-domain approach, exponential scheme	156	0.60
Time-domain approach, Van Leer flux limiter	238	0.91
Time-domain approach, Superbee flux limiter	262	1
Frequency-domain approach	1343	5.13

Table 1: CPU times associated with different time-based approaches and the frequency-based methods for the problem 1.

Resolution methods	CPU time [s]	Relative CPU time (Superbee flux limiter) [-]
Time-domain approach, exponential scheme	143	0.57
Time-domain approach, Van Leer flux limiter	225	0.89
Time-domain approach, Superbee flux limiter	252	1
Frequency-domain approach	1303	5.17

Table 2: CPU times associated with different time-based approaches and the frequency-based methods for the problem 2.