Journal of Quantitative Spectroscopy & Radiative Transfer 93 (2005) 139–150

Journal of Quantitative Spectroscopy & Radiative Transfer

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Theoretical analysis of frequency and time-domain methods for optical characterization of absorbing and scattering media

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Received 1 February 2004; accepted 1 July 2004

Abstract

Measurements of the temporal distribution of radiation transmitted and/or reflected by an absorbing and scattering medium, subject to a very short pulse of light, are likely to provide information on radiative properties of the sample. However, this procedure requires precise solutions of the transient radiative transfer problem as well as appropriate nano/pico response time from detection devices. In this work, a different approach is investigated: the transient radiative transfer problem is solved in the space–frequency domain using a discrete ordinates-finite volume method and is shown to provide accurate results. Recent research work has proposed promising optical diagnostic techniques based on a radiation beam for which intensity is modulated. As a result, a preliminary sensitivity analysis is performed to determine some optimal parameters for the design of frequency-modulation based optical diagnostic techniques. The novelty here also arises from the application of the space–frequency method to media that are not necessarily optically thick.

Keywords: Radiative metrology; Time-resolved technique; Frequency-resolved technique; Discrete ordinates method; Sensitivity analysis; Optimal experiments

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doi:10.1016/j.jqsrt.2004.08.018

Nomenclature

a	anisotropy factor
A	amplitude ($\mathrm{W\,m^{-2}\,\mu m^{-1}}$)
f	frequency (Hz)
H	Heaviside function
$\widehat{I}$	frequency-dependent intensity ($\mathrm{W\,m^{-2}\,sr^{-1}\,\mu m^{-1}}$)
$\bar{I}$	average flux ($\mathrm{W\,m^{-2}\,\mu m^{-1}}$)
k	discrete frequency step
L	path of scattered radiation (m)
S	source term ($\mathrm{W\,m^{-2}\,\mu m^{-1}}$)
X^{norm}	normalized sensitivity coefficient
z	spatial coordinate (m)

Greek letters

α	known parameters vector
η	output vector (measurement)
μ	direction cosine
$\widehat{\omega}$	dimensionless angular frequency (rad)
θ	polar angle (rad)
ϕ	azimuthal angle (rad)
ψ	vector of parameters to be estimated

Subscripts/Superscripts

0	incident collimated beam
$*$	time dimensionless quantity
$\wedge$	frequency-dependent quantity
$'$	other directions
c	collimated component
d	diffuse component
k	refers to a discrete frequency step
L	refers to medium thickness
p	refers to the pulse
R	reflected signal
T	transmitted signal

1. Introduction

Since the original work of Rackmil and Buckius [1], interest for transient radiative transfer has recently increased, mainly because of numerous possible uses of short pulse lasers in a wide variety of engineering and biomedical areas [2]. Optical diagnostic of absorbing and scattering media using temporal distributions of transmittance and reflectance remains a promising application of transient radiative transfer [2].

In a typical transient radiative transfer problem, an absorbing and scattering medium is illuminated by a short laser pulse, whose duration is of the same order of magnitude, or less, than the time required by the radiation to leave the medium [3]. Recently, in order to assess the use of the diffusion approximation for the solution of transient radiative transfer problems, Elaloufi et al. [4] solved the time-dependent radiative transfer equation in the space–frequency domain by use of the discrete ordinates (DO) method. The same approach is retained in the present work. It will be demonstrated that it provides accurate solutions, without physically unrealistic transmitted radiation at early time periods [2].

Moreover, research works in the biomedical area [5,6] proposed a radiative metrology based on a collimated radiation beam whose intensity is modulated in amplitude at one given frequency. Compared to time-domain measurements, the main advantage is that continuous sources and detectors are used in the frequency domain. Although practical limitations related to the frequency modulation can be encountered, frequency-domain measurement techniques constitute very promising alternatives to time-domain technologies. However, since the application has been mainly restricted to the determination of radiative properties of optically thick and highly scattering media, such as biological tissues, the modeling in the frequency domain has been often limited to the use of the diffusion approximation.

Consequently, the objectives of this work are twofold. First, time-domain and frequency-domain methods dedicated to the solution of transient radiative transfer problems are compared. The theoretical formulation is summarized in the first section: the problem under consideration and the associated assumptions are described and the equation of transfer in the frequency domain—called the complex radiative transfer equation (CRTE)—is presented. The method is later applied to typical 1D transient radiative transfer problems and results are compared with those obtained from time-domain solution methods: a Monte Carlo formulation [7] and a DO approach using a Van Leer flux limiter [8]. Time-domain and frequency-domain measurement techniques are then briefly discussed. Some promising features related to frequency modulation approaches are pointed out. The last section of this paper is devoted to the second objective: a preliminary sensitivity analysis is performed to derive some optimal parameters for the design of frequency-based optical diagnostic techniques of absorbing and scattering media.

2. Theoretical formulation

2.1. Time-dependent radiative transfer problem description and assumptions

Throughout this paper, the following assumptions will take place: (1) the sample is a plane-parallel semi-infinite layer of thickness z_L; (2) the slab is composed of an non-emitting, absorbing and scattering homogenous medium, with a unit refractive index; (3) radiative properties are calculated at the central wavelength of the pulse spectral bandwidth and consequently reference to wavelength is omitted in the notations; (4) scattering is assumed to be independent; (5) boundaries are transparent; (6) the slab is subjected to a collimated short pulse radiation at normal incidence (the problem is azimuthally symmetric); (7) a pure transient radiative transfer regime is considered, i.e. the pulse width is less than the characteristic time for the establishment of any other phenomenon [2].

This transient radiative heat transfer problem is schematically described in Fig. 1. A part of the pulsed collimated radiation beam leaves the medium without being deviated, while the other fraction is multiply scattered in all directions. Since the problem deals with collimated irradiation, the most convenient approach is to consider a separate treatment of the diffuse-scattered component (I_d) [9]. Variations of the collimated intensity (I_c) are simply described by a spatial exponential decay and a temporal term originating from the propagation of the pulse [2,3].

The transient radiative transfer equation (TRTE) describes temporal and spatial variations of the diffuse component of the intensity along the direction μ in a participating medium. Using dimensionless time ($t^* = \beta ct$) and optical depth ($\tau = \beta z$) as variables, the TRTE is written as [2,3]

$$\frac{\partial I_\mathrm{d}(\tau,\mu,t^*)}{\partial t^*} + \mu\,\frac{\partial I_\mathrm{d}(\tau,\mu,t^*)}{\partial \tau} = -I_\mathrm{d}(\tau,\mu,t^*) + \frac{\omega}{2}\int_{-1}^{1} I_\mathrm{d}(\tau,\mu',t^*)\Phi(\mu',\mu)\,\mathrm{d}\mu' + S_\mathrm{c}(\tau,\mu,t^*). \tag{1}$$

In the case of a square pulse of duration t_p, the radiation source term S_c, due to the scattering of the collimated intensity, is given by [3]

$$S_\mathrm{c}(\tau,\mu,t^*) = \frac{\omega}{4\pi}\, I_0\, \exp(-\tau)[H(t^*-\tau) - H(t^* - t_\mathrm{p}^* - \tau)]\Phi(1,\mu). \tag{2}$$

2.2. *Radiative transfer equation in the space–frequency domain*

As one of the objectives 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 of the intensity $I(\tau,\mu,t^*)$ [4]:

$$I(\tau,\mu,t^*) = \int_{-\infty}^{\infty} \widehat{I}(\tau,\mu,\widehat{\omega})\exp(\mathrm{i}\widehat{\omega}t^*)\,\mathrm{d}\widehat{\omega}, \tag{3}$$

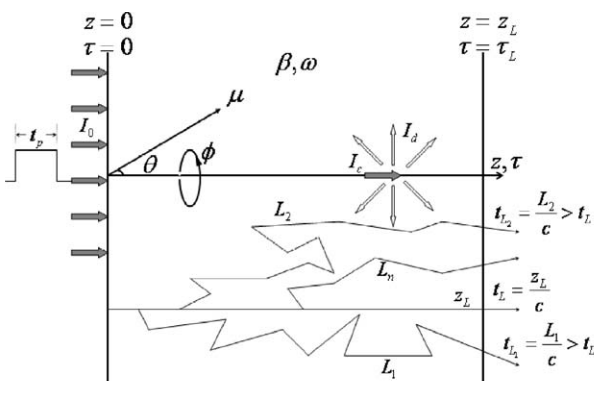


Fig. 1. Schematic representation of a typical transient radiative transfer problem.

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

$$\mu \frac{\mathrm{d}\widehat{I}_{\mathrm{d}}(\tau,\mu,\widehat{\omega})}{\mathrm{d}\tau} = -(1+\mathrm{i}\widehat{\omega})\widehat{I}_{\mathrm{d}}(\tau,\mu,\widehat{\omega}) + \frac{\omega}{2}\int_{-1}^{1} \widehat{I}_{\mathrm{d}}(\tau,\mu',\widehat{\omega})\Phi(\mu',\mu)\,\mathrm{d}\mu' + \widehat{S}_{\mathrm{c}}(\tau,\mu,\widehat{\omega}), \tag{4}$$

where the frequency-dependent intensity $\widehat{I}_{\mathrm{d}}(\tau,\mu,\widehat{\omega})$ and $(1+\mathrm{i}\widehat{\omega})$ are complex numbers. This equation has the form of a steady-state radiative transfer equation (RTE) and is called the complex radiative transfer equation (CRTE). The source term $\widehat{S}_{\mathrm{c}}$ originating from the scattering of the collimated intensity is determined from a particular solution of Eq. (4) without scattering sources [4]

$$\widehat{S}_{\mathrm{c}}(\tau,\mu,\widehat{\omega}) = \frac{\omega}{4\pi}\widehat{I}_0(\widehat{\omega})\exp[-\tau(1+\mathrm{i}\widehat{\omega})]\Phi(1,\mu), \tag{5}$$

where $\widehat{I}_0(\widehat{\omega})$ is obtained from the temporal Fourier analysis of $I_0(t^*)$.

From the above statement, it is concluded that the solution of a transient radiative transfer problem can be obtained by solving a steady-state radiative transfer equation for each frequency contained in the temporal Fourier decomposition of the pulse.

3. Time-domain vs. frequency-domain methods

3.1. Transient radiative transfer solution using a frequency-domain method

A standard discrete ordinates-finite volume method (DO-FV) [9] is used in order to solve the CRTE. The principal steps are summarized hereafter: (1) the temporal Fourier transform of the light pulse is calculated; (2) the CRTE (Eq. (4)) is solved for each angular frequency ($\widehat{\omega}$) contained in the pulse using the DO-FV method; (3) an inverse Fourier transform is applied in order to derive time-dependent variables.

The frequency-domain approach is compared with two time-domain methods: the first is based on a DO approach using a high-order accurate flux limiter as spatial differencing scheme and the second on a Monte Carlo formulation [7]. Flux limiters are correction factors which allow the representation of a sharp gradient with a minimum of numerical diffusion. The Van Leer flux limiter was applied recently for 3D transient radiative transfer problems using a MOL (Method of Lines) solution of DO method [8] and was shown to be very satisfactory. We also developed a DO-FV code applying the Van Leer flux limiter to solve the TRTE; a good agreement with the Monte Carlo formulation was found [7], except at early time period (results not shown). In each of the following test cases, a square pulse with $t_{\mathrm{p}}^* = 1$ and $I_0 = 1$ is considered. The temporal transmittance, defined as the hemispherical flux leaving the medium at the boundary $\tau = \tau_L$, is reported in Fig. 2. Radiative properties of the media are given directly on the figure, where a refers to the anisotropy factor associated to the linear anisotropic phase function $\Phi(\mu',\mu) = 1 + a\mu\mu'$.

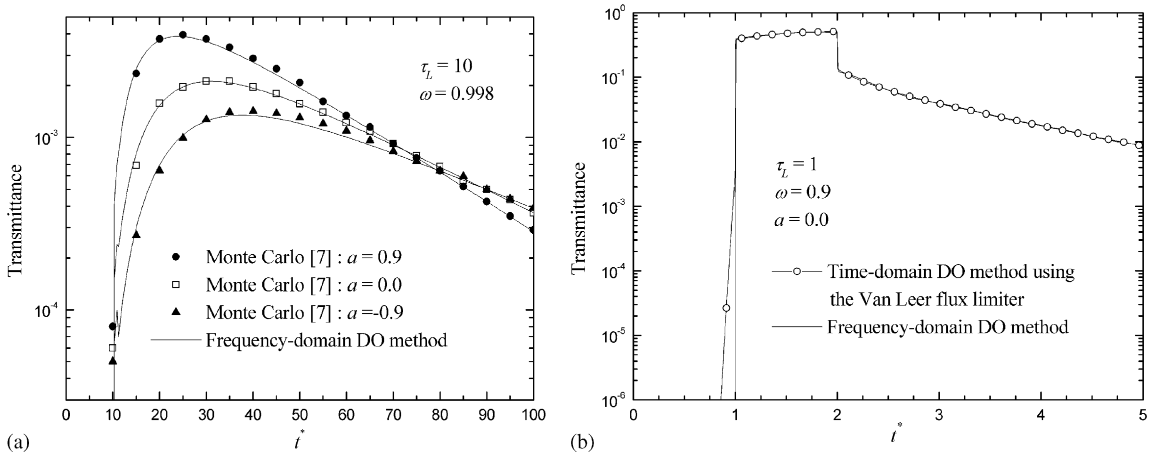


Fig. 2. Comparison between the frequency-domain method and (a) a time-domain Monte Carlo formulation [7]; (b) a time-domain DO method using the Van Leer flux limiter.

The temporal Fourier transform of the square pulse (sinus cardinal function) allows the determination of pulsations $\widehat{\omega}$ (frequencies) for which the CRTE has to be solved. A preliminary parametric analysis permitted the identification of an optimal angular frequency discretization ($\Delta\widehat{\omega}_k = 3 \times 10^{-4}\pi$).

An excellent agreement between the results obtained with the proposed method and those obtained from a Monte Carlo formulation is observed (Fig. 2(a)). The mean relative differences between Monte Carlo and frequency-domain DO method results are 2.35%, 2.57% and 4.47% for anisotropy factors of 0, 0.9 and −0.9, respectively. Moreover, using the same scales for the axes, similar agreement would be found with a standard DO-FV/Van Leer method (not shown). A comparison with the DO-FV solution using a Van Leer flux limiter shows that the frequency-domain method is indeed accurate, even at the early time period (Fig. 2(b)). Here, the minimal dimensionless time requested by the radiation to leave the medium for an optical thickness, $\tau_L = 1$, is $t^*_{\text{min}} = 1$. It can be seen that the transmitted flux, in the case of the DO-FV/Van Leer approach, emerges earlier from the domain. This is not the case for the frequency-domain formulation, where the transmittance begins exactly at $t^* = 1$. These test cases concisely illustrate the efficiency of the frequency-domain approach when applied to solve time-dependent radiative transfer problems.

3.2. Time-domain vs. frequency-domain measurement techniques

Transient radiative transfer solvers are used to analyze the temporal distribution of transmitted and reflected signals for metrological applications. As an example, in biomedical research, time-domain measurement techniques use near-infrared (NIR) pulsed light sources (pulse duration from femto to nanoseconds), because biological tissues partially scatter and absorb radiation at this wavelength range. The temporal distribution of photons transmitted or reflected by the scattering medium carries information on tissues radiative properties. The temporal response extends only over a few nanoseconds. As for examples of detector devices, streak cameras can record radiation fluxes with a temporal resolution of a few picoseconds. However, the main drawback of streak cameras is its high cost [5]. Then time-domain measurement techniques may lead to intrinsic experimental difficulties, mainly because of current detectors limitations.

Research works from the biomedical area [5,6] suggested an alternative radiative metrology in the frequency domain. A schematic comparison between time- and frequency-domain measurement techniques is described in Fig. 3. For the frequency approach, the intensity of a continuous light source (NIR for biomedical applications) is modulated in amplitude at a given frequency (f) and is directed towards the participating medium. The average intensity and the amplitude of transmitted $A_T(f)$ or reflected $A_R(f)$ radiations, and its phase shift related to the incident light, are measured and analyzed to derive radiative properties of the participating medium using an appropriate parameter estimation method. Since continuous light sources are used, detection limitations associated with time-domain techniques are avoided. However, modulation frequencies which have been produced so far are less than a few hundred MHz. Despite this, the technique is currently used for biological tissue imaging and is very promising for other engineering applications.

As stated by Yodh and Chance [5], it is not clear which experimental approach will find most use. For a single measurement, temporal techniques provide more information since a pulse contains many frequencies. However, frequency-domain approaches are technically simpler to develop and essentially give the requested information to infer radiative properties, provided that most relevant discrete frequencies are selected. As a consequence, the next section is devoted to a preliminary sensitivity analysis in an attempt to derive some optimum parameters for the design of such optical diagnostic techniques.

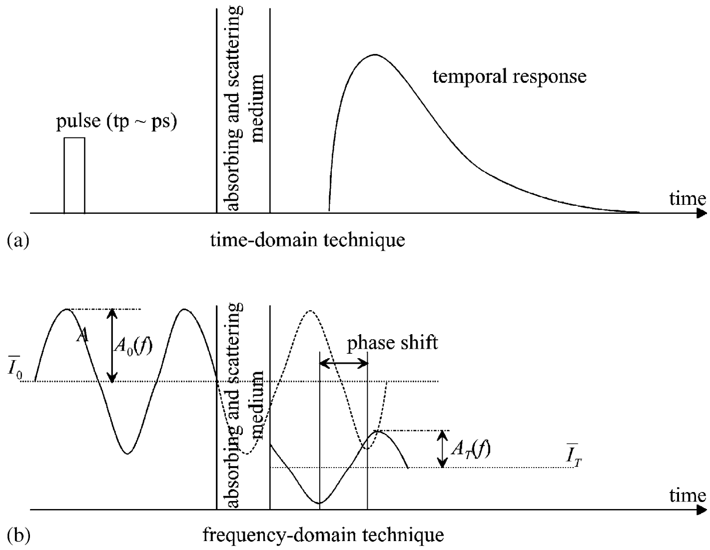


Fig. 3. Time-domain (a) vs. frequency-domain (b) measurement technique principles.

4. Sensitivity analysis in the frequency domain

4.1. Basics of sensitivity coefficients

In parameter estimation problems, the sensitivity analysis plays a key role. In particular, it helps designing optimal experiments through the adjustment of known parameters (input, known properties of the system) so that the output (the measurement of a relevant variable, noise put aside) will be as sensitive as possible to the parameters we want to estimate [10]. The normalized sensitivity coefficient

$$X_{\psi_i}^{\mathrm{norm}}[\eta](\alpha,\psi) = \left|\frac{\partial\eta(\alpha,\psi)}{\partial\psi_i}\right| \frac{\psi_i}{\eta(\alpha,\psi)}, \tag{6}$$

provides the relative variation of an output (η) associated to a relative variation of one parameter (ψ_i) of the system, when all other parameters (known α or to be estimated $\psi_{j,j\neq i}$) are fixed. Generally, parameter estimation is conceivable when normalized sensibility coefficients are greater than 0.1, difficult when they are between 0.01 and 0.1, and very difficult, or even impossible, when they are less than 0.01. These criteria are approximate since they depend on the detector sensitivity.

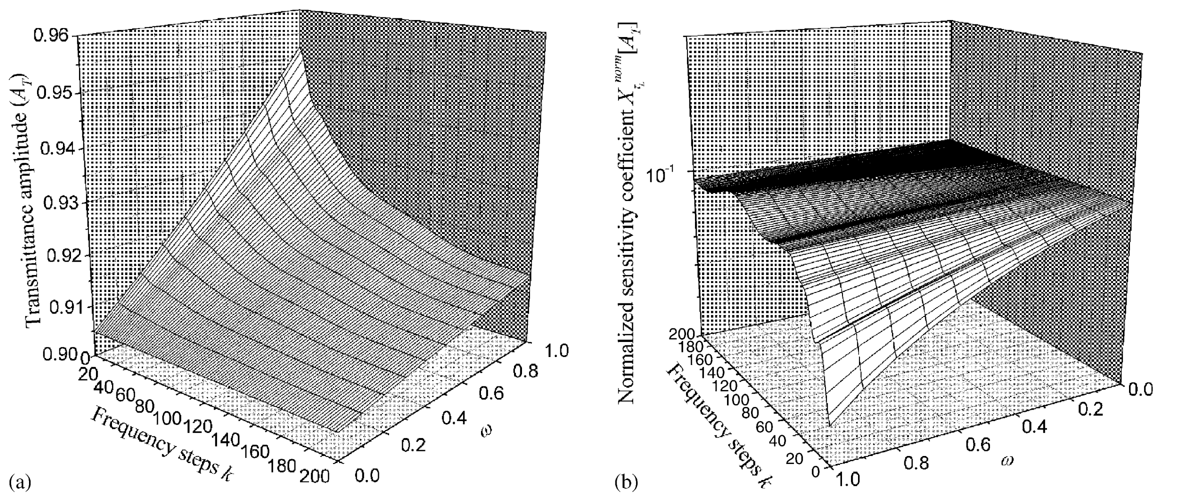
4.2. Results and discussion

In this study, parameters (ψ_i) for which normalized sensitivity coefficients are evaluated are the scattering albedo ω and the sample optical thickness τ_L. As for output values (η), transmittance and reflectance amplitudes (A_{T} and A_{R}, respectively) are analyzed, as a function of the time dimensionless pulsation $\widehat{\omega}$, which varies from 0 to 20π with a constant step ($\widehat{\omega}_k = k\pi/10$). It must be mentioned that to ensure generality, the analysis is performed with an angular frequency derived from the dimensionless time $t^* = \beta ct$. This implies that for each practical application, the optimal set-up will be obtained from frequency results (determined according to $f = \tau_L c\widehat{\omega}/(2\pi z_L)$). However, to obtain this frequency, an iterative approach is required to fix the sample thickness (z_L) and its optical thickness (τ_L). Amplitudes and normalized sensitivity coefficients are presented on selected 3D plots for a unit amplitude of the modulated incident light source ($A_0 = 1$), as a function of the scattering albedo ω and of the pulsation $\widehat{\omega}$. Three optical thicknesses ($\tau_L =$ 0.1, 1 and 10) are considered and scattering is assumed to be isotropic ($a = 0$).

As a preliminary discussion, it should be noted that multiple scattering effects cannot be observed for transmittance amplitudes when the source modulation frequency f is much larger than a critical frequency ($f_{\mathrm{crit}} = c\sigma_{\mathrm{s}}$) corresponding to the reciprocal of the average time between consecutive scattering events [11]. In other words, it means that the transmittance amplitude hardly depend on f when $f \gg f_{\mathrm{crit}}$ and simply results from the Beer law ($A_{\mathrm{T,crit}} = \mathrm{e}^{-\tau_L} A_0$). In terms of dimensionless angular frequency, the critical value is expressed as a function of the scattering albedo ($\widehat{\omega}_{\mathrm{crit}} = 2\pi\omega$). For scattering albedos of 0.1, 0.5 and 1, critical dimensionless pulsations are, respectively, 0.63, 3.14 and 6.28, corresponding to frequency steps (k) equal to 2, 10 and 20, respectively.

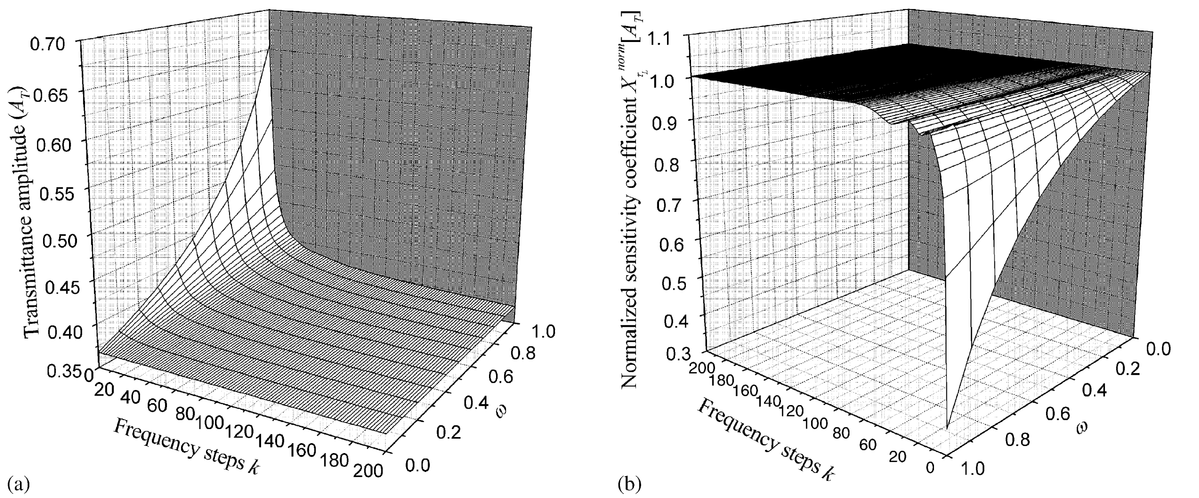
4.2.1. Sensitivity of amplitudes to the optical thickness

As explained above, it is observed in Figs. 4(a) and 5(a) that the transmittance amplitude A_{T} does not practically vary with k for frequency steps larger than the critical values (the amplitude scale spreads only over a few hundredths). For optical thicknesses of 0.1 and 1, only the transmittance amplitude A_{T} has been considered since the reflectance amplitude A_{R} is generally too small (except at low frequencies for highly scattering media). Therefore, only normalized sensitivity coefficients of A_{T} to the optical thickness are presented in Fig. 4(b) for $\tau_L = 0.1$, where it is shown that when the scattering albedo augments, frequency has to be slightly increased to keep the sensitivity to a maximal and acceptable value of about 0.1. This can be partially



(a) (b)

Fig. 4. (a) Transmittance amplitude $A_{\mathrm{T}}(\widehat{\omega}_k, \tau_L = 0.1, \omega)$; (b) normalized sensitivity coefficient $X_{\tau_L}^{\mathrm{norm}}[A_{\mathrm{T}}](\widehat{\omega}_k, \tau_L = 0.1, \omega)$.



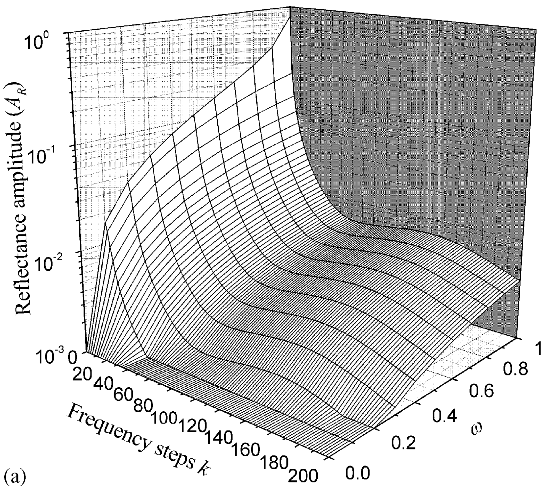
(a) (b)

Fig. 5. (a) Transmittance amplitude $A_{\mathrm{T}}(\widehat{\omega}_k, \tau_L = 1, \omega)$; (b) normalized sensitivity coefficient $X_{\tau_L}^{\mathrm{norm}}[A_{\mathrm{T}}](\widehat{\omega}_k, \tau_L = 1, \omega)$.

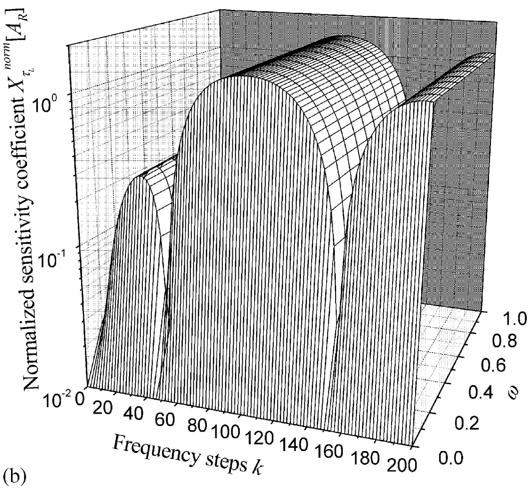
explained by the fact that A_T essentially depends on the optical thickness when modulation frequencies are much larger than critical frequencies. It is worth mentioning here that very high frequencies are not suitable as they imply that the detection device would not detect the amplitude or such a modulated laser even does not exist. In Fig. 5(b), the same sensitivity coefficient for an optical thickness of 1 is found to be nearly constant and equal to unity, except at very low frequencies for highly scattering media, where sensitivity coefficients are still acceptable (minimum of approximately 0.35). For optically thick media ($\tau_L = 10$, not shown), the transmittance amplitude A_T is becoming too weak, except for highly scattering media ($\omega \to 1$) at low frequencies. Therefore, the reflectance amplitude A_R is considered (Fig. 6(a)). It can be seen that the reflectance amplitude is of course an increasing function of albedo and shows a global downward trend when frequency increases. This can be partially explained by the fact that for frequencies higher than the critical frequency f_{crit}, multiple scattering effects cannot be observed for the oscillating component of radiation intensity. Fluctuations of A_R need further investigation in order to explain the exhibited variations of normalized sensitivity coefficients (Fig. 6(b)). However, it is possible to affirm that the optimal zone for A_R is for the low-frequency range ($15 < k < 40$) and for scattering albedos higher than 0.3 if we consider both sensitivity and detection requirements.

4.2.2. Sensitivity of amplitudes to the scattering albedo

The normalized sensitivity coefficients for A_T are depicted in Figs. 7 ($\tau_L = 0.1$) and 8 ($\tau_L = 1$). For optically thin media, sensitivities are still acceptable for low frequencies and scattering albedos larger than 0.2. Increasing the optical thickness to unity makes this sensitivity increase, particularly for low angular frequencies and for highly scattering media. As aforementioned, this behavior can be explained by the fact that multiple scattering effects cannot be observed for transmittance amplitudes when modulation angular frequencies are much greater than the critical value $\widehat{\omega}_{crit}$; then, largest values of normalized sensitivity coefficients are found for low modulation frequencies and highly scattering media. Finally, A_R is highly sensitive to the scattering albedo for



(a)



(b)

Fig. 6. (a) Reflectance amplitude $A_R(\widehat{\omega}_k, \tau_L = 10, \omega)$; (b) normalized sensitivity coefficient $X_{\tau_L}^{norm}[A_R](\widehat{\omega}_k, \tau_L = 10, \omega)$.

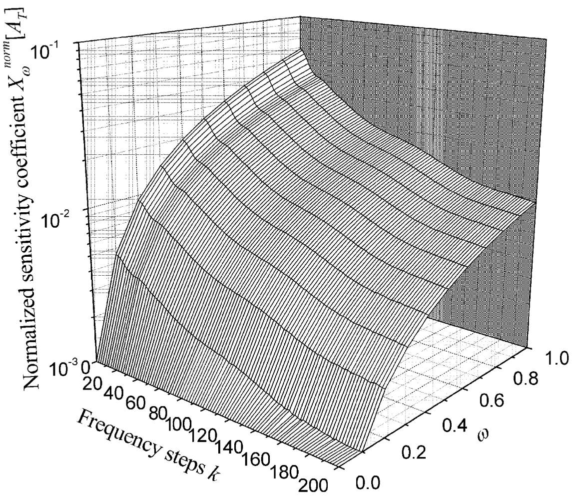


Fig. 7. Normalized sensitivity coefficient $X_\omega^{\mathrm{norm}}[A_\mathrm{T}](\widehat{\omega}_k, \tau_L = 0.1, \omega)$.

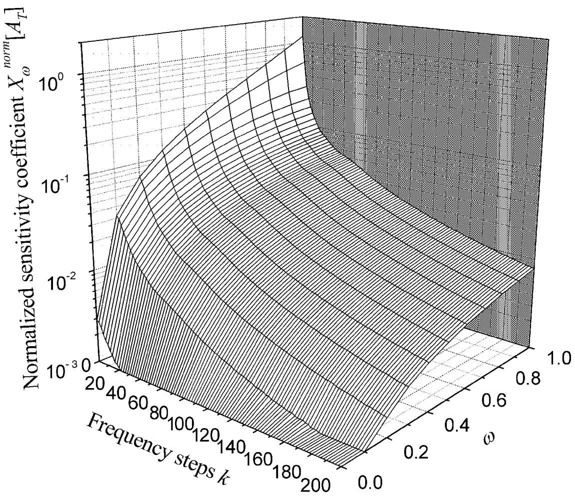


Fig. 8. Normalized sensitivity coefficient $X_\omega^{\mathrm{norm}}[A_\mathrm{T}](\widehat{\omega}_k, \tau_L = 1, \omega)$.

an optically thick medium ($\tau_L = 10$), regardless of frequency and provided scattering albedo is higher than a few percents (not shown).

This preliminary analysis, giving optimal parameters in terms of output and dimensionless angular frequency choices for absorbing and isotropic scattering media, has to be extended in future developments to the study of phase shifts and average fluxes and/or demodulation data (obtained from source and detected amplitude on average fluxes ratios $((A_\mathrm{T}/\overline{I}_\mathrm{T})/(A_0/\overline{I}_0)$ in Fig. 3)).

5. Conclusion

The transient radiative transfer problem for absorbing-scattering media has been solved in the frequency-dependent domain by use of a standard discrete ordinates-finite volume method. It has been shown that solutions are accurate, without physically unrealistic transmitted radiation at early time periods. It has also been pointed out that frequency-domain measurement techniques, i.e. using a radiation beam whose intensity is modulated in amplitude at a given frequency, are very promising to characterize participating media, for a wide spectrum of engineering applications. In order to determine optimal parameters to design such techniques, sensitivities of reflectance and transmittance amplitudes to the optical thickness and to the scattering albedo have been analyzed. The sensitivity analysis must be developed further for other configurations such as anisotropically scattering media and by applying it to the phase shift and to the average intensity outputs. It will be thus possible to provide accurate criteria such as best modulation frequencies for measurement setups optimal design and to assess the identification of radiative properties.

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