Proceedings of
2002 ASME International Mechanical Engineering Congress and Exposition
November 17–22, 2002, New Orleans, Louisiana

IMECE2002-33903

EXPERIMENTAL AND NUMERICAL STUDIES OF SHORT PULSE PROPAGATION IN MODEL SYSTEMS

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ABSTRACT

In this paper experimental and numerical studies of short-pulsed lasers propagation through scattering and absorbing media are investigated. Experimental results of a 60 ps pulse laser transmission in tissue phantoms are presented and compared with Monte Carlo simulation. Good agreement between the Monte Carlo simulation and experimental measurement is found. Three models are developed for the simulation of short pulse transport. Benchmark comparisons among the Monte Carlo, transient discrete ordinates method and transient radiation element method are conducted.

INTRODUCTION

The study of short pulse laser propagation through highly scattering media has received considerable recent interest mainly due to its applications in biomedical imaging such as optical tomography, remote sensing of oceans and atmospheres, laser material processing of microstructures, etc. The unique features of short pulses of radiation and the characteristics of their interaction with participating medium make them a valuable research tool. Firstly, short pulse monochromatic radiation can be easily created and accurately detected with high spatial, temporal, and signal resolution. Secondly, the distinct feature of the technique is the multiple scattering induced temporal signature that persists for time periods greater than the source pulse duration and is a function of the scattering-absorbing properties of the medium and the location in the medium where the properties undergo changes. This temporal variation of reflected and transmitted signals is of significance in visualization, imaging and modeling.

The modeling of multi-dimensional radiative transfer is usually a formidable task even in steady state since it involves solving integro-differential equations. The addition of the transient term in transfer equation further increases the complexity of modeling. There are mainly two categories in the modeling of transient radiative transfer, i.e., the stochastic method and the deterministic method. The stochastic Monte Carlo (MC) method is usually adopted for the simulation of short pulse laser propagation since it avoids the handling of the complicated integro-differential relationship, is flexible to deal with realistic physical conditions, and is algorithmically simple (Flock *et al.*, 1989; Hasegwa *et al.*, 1991; Guo et al., 2000).

The shortcomings of the Monte Carlo method are that it is time-consuming and that the results are subject to statistical error due to practical finite samplings. In brain tumor diagnosis, for example, the nondimensional optical thickness of the tissue is usually over 100, the ballistic component of the laser beam passing through the tissue is then in the order of exp(-100), which is very small to be captured precisely by the Monte Carlo method even with the use of a huge number of samplings.

In contrast, the deterministic method does not suffer such defects. The most commonly used model in biomedical field is the diffusion theory, in which the light is assumed to be multiply scattered and the radiative transfer equation is simplified into a diffusion or parabolic equation for the solid-angle independent fluence (Yamada, 1995; Madsen et al., 1992; Ishimaru, 1987). Time-resolved experiments on tissues have shown that such diffusion-based analyses are accurate for thick samples but fail to match experimental data for thin samples (Yoo et al., 1990; Yodh and Chance, 1995). In addition, it fails to predict adequately the radiative transport in the region near light incidence and near surface boundaries.

In the solution of complete time-dependent radiative transfer equation (RTE), some previous studies have solved the one-dimensional hyperbolic transient RTE, but with a constant strength at the boundaries for a conservative medium using adding-doubling method (Rackmil and Buckius,1983). Hyperbolic formulation using P_1 model was proposed by Ishimaru (1987). Kumar *et al*. (1996) employed the P_1 model to solve the hyperbolic one-dimensional transient transfer equation. Mitra et al. (1997) further developed the P_1 model to two-dimensional rectangular enclosure. More recently, Mitra and Kumar (1998) examined discrete ordinates method, P_1 and P_3 models, diffuse approximation, and two-flux method for short pulse laser transport in one-dimensional planar medium, and found that the discrete ordinates method predicts more accurate transient results. After that, Tan and Hsu (2000) and Wu and Wu (2000) separately developed integral formulation for transient radiative transfer in one- and two-dimensional geometries, but it has not yet been applied to short pulse laser transport. Guo and Kumar (2001a) developed transient radiation element method for 1D plane-parallel systems.

Few studies are addressed to multi-dimensional short pulse radiation transport through turbid media. Guo and Kumar (2001b, 2001c) have formulated transient discrete ordinates method for multi-dimensional geometries. Guo (2001) developed 3D transient radiation element method.

The objective of the work is to study the characteristics of ultrashort laser pulses radiation transport in multi-dimensional geometries containing scattering and absorbing turbid medium. The research goal is to develop adequate transient radiative transfer models for utilization as tools in simulation based engineering and biomedical applications. Experimental study of short pulse laser transport through scattering media is performed for the validation of the numerical models. Comparisons between experimental measurement and numerical modeling are given. Benchmark comparisons among the models are also made.

NOMENCLATURE

c = speed of light
g = asymmetric factor
G = incident radiation
I = radiation intensity
$\mathbf{r}$ = spatial location vector
$\hat{\mathbf{s}}$ = unit vector of direction
t = time
t_p = pulse width
x,y,z = coordinates
μ = direction cosine
q = polar angle
ϕ = azimuthal angle
Φ = scattering phase function
σ_a = absorption coefficient
σ_e = extinction coefficient
σ_s = scattering coefficient
ω = scattering albedo or solid angle

Subscript

b = blackbody
c = collimated

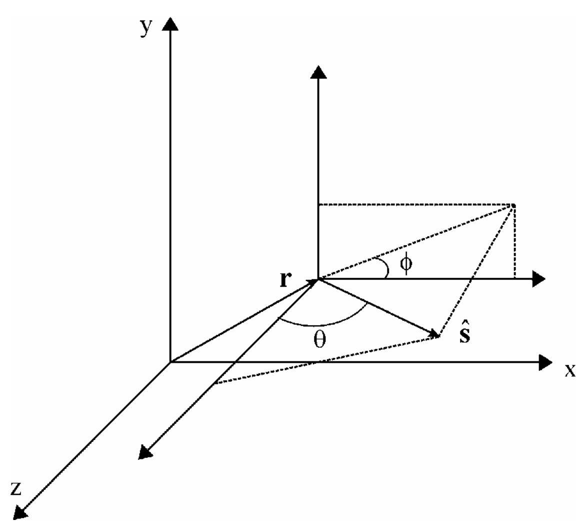

Figure 1. Schematic of the coordinate system.

MATHEMATICAL MODELS

The physical problem under consideration is that ultrashort laser pulses transport through three-dimensional turbid media such as biological tissues. The scattering and absorption properties in turbid media govern the transport of laser. Because of the inhomogeneity of tissues, the solution of Maxwell's equations, which might otherwise be used to model the propagation of light in tissues, is not feasible (Wilson and Patterson, 1986). However, upon ignoring polarization and diffraction effects, the transport of photons through random media (such as tissues) may be modeled as neutral particle transport (Ishimaru, 1987). The radiative transfer equation is then the “exact” model to describe laser radiation transport in scattering, absorbing and emitting turbid media and it can be written as (Siegel and Howell, 1992; Modest, 1993):

$$\frac{1}{c}\frac{\partial I(\mathbf{r},\hat{\mathbf{s}},t)}{\partial t}+\hat{\mathbf{s}}\cdot\nabla I\ (\mathbf{r},\hat{\mathbf{s}},t)=-(\sigma_a+\sigma_s)I(\mathbf{r},\hat{\mathbf{s}},t)$$
$$+\sigma_a I_b(\mathbf{r},\hat{\mathbf{s}},T,t)+\frac{\sigma_s}{4\pi}\int_{4\pi}I(\mathbf{r},\hat{\mathbf{s}}',t)\ \Phi(\hat{\mathbf{s}}'\to\hat{\mathbf{s}})\,\mathrm{d}\,\omega+S(\mathbf{r},\hat{\mathbf{s}},t) \quad \textbf{(1)}$$

where I is the radiation intensity, ∇ the gradient operator, t the time, $\mathbf{r}$ the spatial location vector, $\hat{\mathbf{s}}$ the unit vector in the direction of intensity, S the source term, σ_a the absorption coefficient, σ_s the scattering coefficient, ω the spatial solid angle, $\Phi(\hat{\mathbf{s}}'\to\hat{\mathbf{s}})$ the scattering phase function. Figure 1 shows the schematics of the coordinate system under consideration.

The radiation intensity in the medium can be separated into a collimated component and one other remaining part when collimated laser irradiation is considered. If the collimated intensity is I_c then I is the remaining part which can be described by Eq. (1) and the collimated intensity due to incident laser pulse transport forms the source for I. The source term S in Eq. (1) can then be expressed as

$$S=\frac{\sigma_s}{4\pi}\int_{4\pi}I_c(\mathbf{r},\hat{\mathbf{s}}',t)\,\Phi\ \ (\hat{\mathbf{s}}'\to\hat{\mathbf{s}})d\omega \quad \textbf{(2)}$$

where I_c is the collimated component of the incident laser beam.

The radiative heat flux vector $\mathbf{q}$ is expressed as

$$\mathbf{q}(\mathbf{r},t)=\int_{4\pi}I(\mathbf{r},\hat{\mathbf{s}},t)\,\hat{\mathbf{s}}\,d\omega \quad \textbf{(3)}$$

The flux in scalar form is

$$q_i=\mathbf{q}\cdot\hat{\mathbf{e}}_i=\int_{4\pi}I\,\hat{\mathbf{s}}\cdot\hat{\mathbf{e}}_i\,d\omega \quad \textbf{(4)}$$

where $\hat{\mathbf{e}}_i$ is the unit vector along the i-th axis. The incident radiation G is defined as

$$G=\int_{4\pi}I\,d\omega \quad \textbf{(5)}$$

The divergence of radiative heat flux in gray medium is defined as

$$\nabla\cdot\mathbf{q}=\sigma_a(4\sigma T^4-G) \quad \textbf{(6)}$$

where σ is the Stefan-Boltzmann constant and T is the medium temperature which is usually treated as “zero” in the present investigation (cold medium approximation).

The scattering and absorption of radiation by single absorbing spheres can be described by Mie theory (Bohren and Huffman, 1983). Since the Mie phase function is complicated in form and difficult to use in practical radiative transfer analysis, the Henyey-Greenstein (HG) phase function is a reasonable approximation in highly scattering media (Siegel and Howell, 1992) and in biological tissues (Motamedi et al., 1989; Flock et al., 1989). The HG scattering phase function is expressed by

$$\Phi(\mu)=\frac{1-g^2}{(1+g^2-2g\mu)^{3/2}} \quad \textbf{(7)}$$

where g is a dimensionless asymmetry factor that can vary between –1 and 1. This approximate has been shown to produce good agreement with complete Mie scattering calculations (Siegel and Howell, 1992). The advantages of the use of HG phase function are offered in reducing the complexity of the computations and that it introduces only one controlling parameter, namely g, the asymmetry factor.

The incident laser intensity has temporal and spatial Gaussian profiles and can be expressed as

$$I_c(r,t)=I_1\exp\left[-4\ln 2\times\left(t/t_p-0.5\right)^2\right]\times\exp\left(-r^2/\nu^2\right) \quad \textbf{(8)}$$

where t_p is full width at half maximum (FWHM), generally termed as pulse width, and $\nu^2/2$ is the spatial variance of the Gaussian incident beam.

There are mainly two categories in the modeling of transient radiative transfer, i.e., the stochastic method and the deterministic method. The stochastic Monte Carlo method is introduced by Guo et al. (2000) for the simulation of short pulse laser transport in turbid media. Among the deterministic methods, transient discrete ordinates method (TDOM) and transient radiation element method (TREM) are employed in this study. The TDOM method has been formulated by Guo and Kumar (2001b) for two-dimensional geometries and by Guo and Kumar (2001c) for three-dimensional cases. The TREM has been developed by Guo and Kumar (2001a) for plane-parallel systems and formulations for three-dimensional configurations are given by Guo (2001). In the present study, the formulations concerning the three models are not repeated again.

EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 2. The apparatus is composed of three primary parts: a short pulse laser source unit, a sample cell, and an ultrafast detecting unit. Laser pulses of 60 ps at a repetition rate of 76 MHz were generated from frequency-doubled Nd^{+3}:YAG mode-locked laser. The beam diameter is 1.3 mm. A beamsplitter diverts 10% of the 532 nm, 60 ps, 76 MHz laser pulse train to a power meter to monitor the intensity of the incident pulse. The sample is mounted, stationary, in the center of a rotary stage, with an optical fiber mount attached to the rotary platen. The scattered light is collected by an approximately 100 micron optical fiber whose angular position relative to the incident laser beam’s axis can be accurately varied by rotating the mount, enabling monitoring of the angular distribution, intensity and temporal profile of the scattered light by a Hamamatsu OOS-01 optical oscilloscope. The halfwave plate/polarizer combination controls both polarization and the incident power on the sample. An adjustable mask with a circular aperture placed on the output face of the sample limits the area whose scattered output is monitored.

The sample is made to model similar optical properties of artificial biological tissues. In the process of making sample, amorphous silica micro-spheres of very regular shape, diameter

1000nm +/- 10%, were dispersed in styrene, which was then polymerized. These cast solids, typically about 0.81% v/v loaded in the present study, have the advantage of flexibility of shape, minimal number of interfaces to account for in modeling, and lack of convective motion from localized heating due to absorption. Absorption is negligible in the polymer matrix for visible wavelengths, and can be adjusted by mixing dyes into the styrene before polymerization.

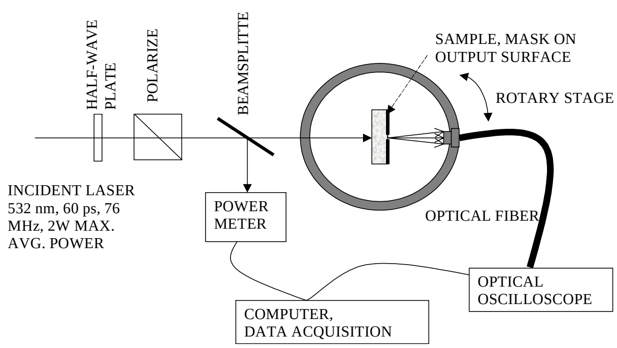

Figure 2. Experimental setup.

The refractive index of the matrix polystyrene is 1.59 at the wavelength of 589 nm (Mark, 1996). The bulk refractive index of the silica spheres is 1.458 at the wavelength of 589 nm (Mark, 1996), while a value of 1.417 was reported at the wavelength of 550 nm for individual silica bead by Firbank et al. (1995). The wavelength of laser used in the experiment is 532 nm. Based on these data, the optical properties of the sample can be calculated using the Mie theory (Bohren and Huffman, 1983). In the calculation, a program supplied in the textbook (Bohren and Huffman, 1983) was employed. Henyey-Greenstein phase function is considered. The calculated optical properties as well as the asymmetric factor for the HG phase function are listed in Table 1. It is seen that the scattering properties of the sample is strongly influenced by the refractive index of the scattering particles. Consequently, the simulation results of short pulse laser transport will be affected.

Table 1. Optical properties of sample in experiment.

R.I. of Silica	σ_s (mm^{-1})	σ_a (mm^{-1})	g
1.417	17.052	0.0	0.96276
1.458	11.309	0.0	0.96649

RESULTS AND DISCUSSION

Experimental measurements and comparison

At first, the sample is made of a block with the thickness in the laser optical axis is 14.4 mm. Other two dimensions are 49 mm × 47.3 mm. Measurement were performed at the azimuthal angles of 0, 30, 50, and 70 degree, respectively. In Fig. 3, the experimental results are presented and compared with the three-dimensional Monte Carlo simulation. The experimental results are shown only for 0 and 30 degree detectors and the amplitudes are normalized. The difference of normalized temporal transmittance between different angles is slight comparing with the measurement uncertainty. In experiment, we found that the uncertainty becomes larger with the increase of the detector azimuthal angle. This is because the magnitude of the transmitted intensity becomes smaller. We found that no matter at what angle the detector is, there exists a plateau in the temporal transmittance at the time period of 1000 ps < t < 1100 ps. At several trials, such a phenomenon did not disappear. It might be caused by the re-entry of backscattered pulse.

In the study of MC method (Guo, 2001), we have shown that the temporal shapes of the transmitted pulse at different azimuthal angles are very similar for diffuse medium, so that a hemispherical transmittance may be used to represent the transmittance detected at specific angle. Experimental results here also show such a behavior. In Fig. 3, the hemispherical results for the MC simulation are presented. In addition, the diameter of detector (optical fiber) in the experiment is only 100 microns and it is located 30 mm away from the output surface. It is very difficult to predict accurately the temporal behavior in a very small detector using the MC method. Even a huge number of samplings is used, the uncertainly of the MC results is severe. Thus, the diameter of detector used in the MC simulation is 1.5 mm.

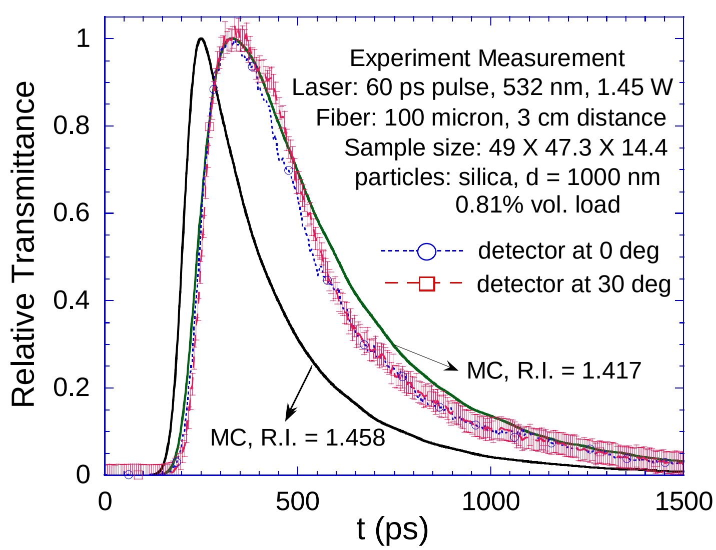

Figure 3. Experimental measurements and comparison with MC simulations for a 60 ps laser pulse passing through a

block. Error bars are plotted for the measurement at detector of 30 deg.

In Fig. 3, it is seen that the predicted temporal profile of transmittance matches closely the experimental measurement in the case of R.I = 1.417 (silica micro spheres). While in the case of R.I = 1.458, the MC predicted transmitted pulse width is obviously narrower than that of the experimental measurement. Since the volume load of silica micro spheres in the present experiment is 0.81%, one should not use the bulk value of refractive index. Thus, R.I = 1.417 is the proper value for the individual silica micro-beads. The measurement of temporal transmittance might be a promising way for measuring refractive index of individual particles. The slight mismatch between prediction and measurement in the range of $500 < t < 1000$ ps can be explained by the different detector radii used in experiment and simulation. Previous research has revealed that the transmitted pulse width in larger radial position is wider.

In Fig. 4 experimental validation is further performed for a sample of cylinder shape. The radius of the cylinder sample is 32.9 mm. The thickness of the cylinder in the optical axis is 19.95 mm. Laser pulse is normally incident at the center of cylinder surface. Error bars are plotted for the measurement at 20 deg detector. Again it is seen that the temporal shapes of the transmitted pulses at different azimuthal angles are very similar. The MC results match the experimental measurement very well.

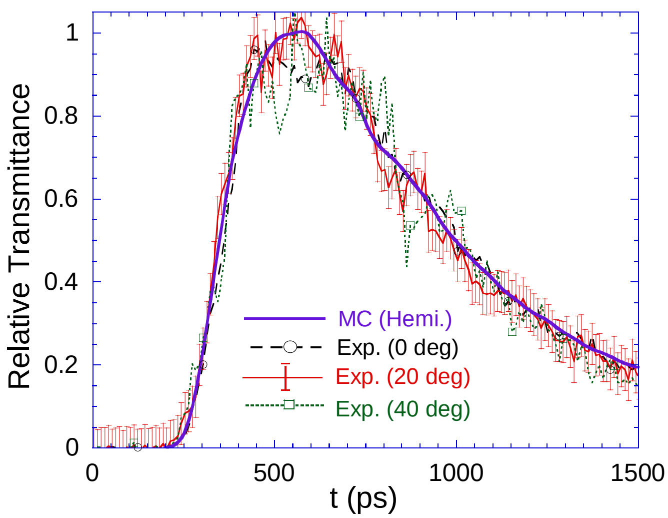

Figure 4. Experimental measurements and comparison with MC simulations for a 60 ps laser pulse passing through a cylinder.

Benchmark comparisons among models

The benchmark problem is chosen as laser irradiation on a cube with unity side length. The optical thickness of the cube is also unity. The medium is cold and purely scattering. The wall with incidence of laser irradiation is called as hot wall and the irradiation is assumed to be diffuse. The rest five walls are cold and black. Since the optical thickness is not very thick, the MC calculation is fast. Therefore, the ray emission number for each MC run is 10^8, and the result is an average of 20 different runs in the benchmark computations. In the TDOM calculations, S_{16} is employed and the cube is divided into a cubic grid of 25×25×25. The size of time step is $\Delta t^* = 0.005$. In the TREM simulation, the cubic radiation element grid is 19×19×19 and the ray emission number for each element is 2141. The CPU time expended in simulation depends on the modeling mechanism. In the MC method, the number of emitted photons, the optical thickness and scattering albedo of the media, and the clock frequency of the CPU are the major factors that affect the CPU time. While in the deterministic methods, such as the TDOM and TREM, the main influential parameters are the grid size and time step employed in the calculation, the memory and cache performance of the CPU. In addition, the radiative transfer information in the whole field can be obtained simultaneously by the deterministic methods, while the current MC method only predicts the reflectance and transmittance information at several specific detector positions and this can save a lot of CPU time. When a Dell PC with one Pentium III Xeon 500 MHz CPU and 512 MB memory is employed for the benchmark calculations, the CPU time is approximately 10 hrs for the MC method, 18 hrs for the TDOM method and 2 hrs for the TREM method, respectively. The relative statistical uncertainty for the MC results is less than 1%.

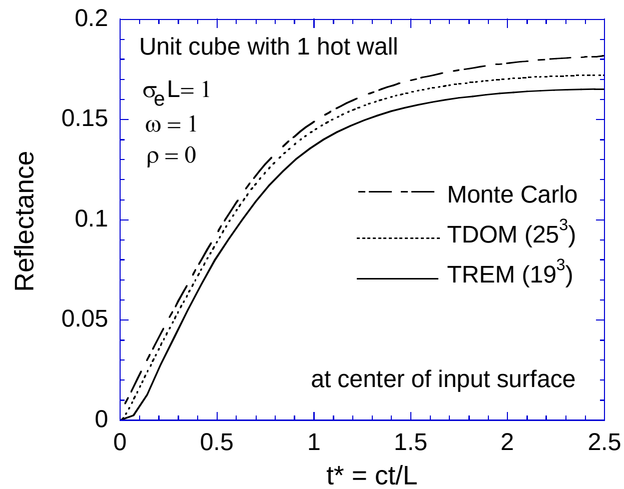

Figure 5. Comparison of the three models for the prediction of temporal reflectance at the input surface center in the benchmark problem subject to a unit laser irradiation from time $t = 0^+$.

Figure 5 shows the comparison of the three models for the prediction of temporal reflectance in the benchmark problem subject to a unit laser irradiation from time $t = 0^+$. It is seen that the temporal behaviors predicted by the three models are very similar. There is obvious difference when the value is concerned. However, the difference of radiation heat transfer between different models is usually high even in steady state. Such a difference in Fig. 5 is relatively small and the accuracy for the three models is acceptable. For the prediction of reflectance, the TDOM matches the MC better than the TREM. One reason could be that the time step in TREM is usually much larger than the time step in TDOM.

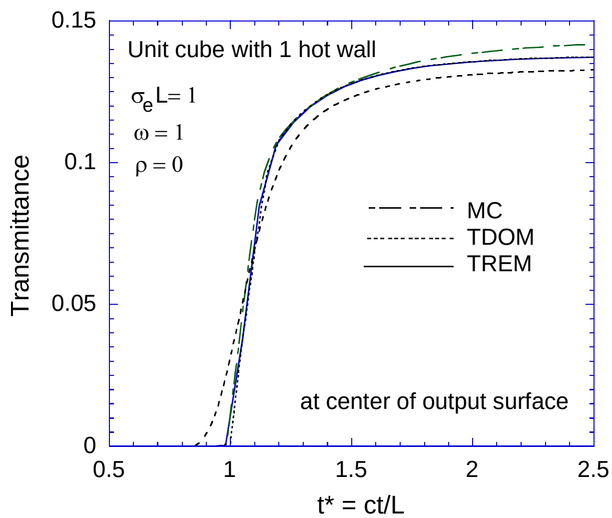

Figure 6. Comparison of the three models for the prediction of temporal transmittance at the output surface center in the benchmark problem subject to a unit laser irradiation from time $t = 0^+$.

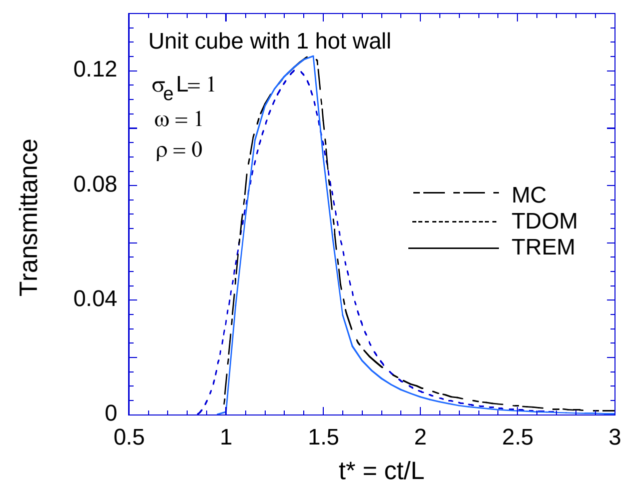

Figure 7. Comparison of the three models for the prediction of temporal transmittance at the output surface center in the benchmark problem subject to a square pulse laser irradiation with pulse duration $t_p^* = 0.5$.

The comparison of the three models for the prediction of temporal transmittance is illustrated in Fig. 6 for the benchmark problem subject to a unit laser irradiation from time $t = 0^+$. Here, the numerical diffusion and false radiation propagation is clearly observed for the TDOM. The TREM result approaches the MC prediction very well in the whole transient time domain. Except at the early time stage, the prediction by TDOM is reasonably accurate as compared with those predicted by the MC and TREM methods.

As we know, the temporal responses for the benchmark problem subject to a pulse laser can be integrated from the previous unit irradiation results in Figs. 5 and 6 through the use of Duhamel's superposition theorem. Here we only present a simple comparison of the three models for a pulsed laser irradiation. Figure 7 shows the comparison of the three models for the prediction of temporal transmittance in the benchmark problem subject to a pulsed laser irradiation. The dimensionless incident laser pulse width is $t_p = 0.5$, and it is a square pulse. It is seen that the TREM matches closely the MC simulation. The transmitted pulse shape predicted by TDOM also approaches the other two pulses in most of the time region.

SUMMARY

Experimental measurement of short laser pulse transport through turbid media is performed. The measured results are compared with the Monte Carlo simulations. Good agreements are found between the measurement and simulation. Both experimental and numerical studies demonstrate that, when the optical thickness of the medium is considerably large, the difference of temporal shapes (except amplitude) between different azimuthal angles is slight. In other words, the temporal profile of the transmitted pulse is not a function of the output azimuthal angle for diffuse media. This implies that it might be reasonable to simulate the transmitted pulse at specific detector angles using the hemispherical results.

The use of an accurate value of the refractive index of the scattering particles is very important in simulation. Even an error of several percent will change the scattering properties, and consequently change the prediction of the temporal behavior of pulse transfer. For the present experimental sample, the choice of refractive index of the silica micro sphere should base on its individual microsphere property. The use of a bulk refractive index leads to an obvious erroneous prediction of the optical properties as well as the temporal signals. Since the laser transport is strongly related to the microstructure of the medium, short pulse laser technique can be used to evaluate the properties of the medium in a microscale standpoint, such as the evaluation of refractive index of microspheres.

The benchmark study of the three models shows that the three different methods predict similar transient radiation transfer. In the prediction of transient reflectance, the TDOM approaches the MC better than the TREM. While the TREM matches the MC better than the TDOM in the prediction of temporal transmittance profile. The accuracies of the three models are generally acceptable.

ACKNOWLEDGMENTS. S. Kumar and B. Garetz acknowledge partial support from the Sandia-NSF joint grant AW-9963 administered by Sandia National Lab, Albuquerque, Dr. Shawn P. Burns, Project Manager. Z. Guo acknowledges partial support from the Charles and Johanna Busch Memorial Fund Award at Rutgers, The State University.

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