MODELING OF ULTRAFAST LASER TRANSPORT AND APPLICATIONS

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ABSTRACT
Recent advances in biomedicine involving the use of ultrafast pulsed laser technology, including such applications as optical tomography and plasma-mediated ablation, have mandated the necessity of accurately modeling ultrafast propagation of radiant energy through solution of the time-dependent equation of radiative transfer. Numerical modeling can be implemented as a realistic alternative for situations where ultrafast laser experimentation is either complicated or expensive in nature. In this review, advances in the computational modeling of ultrafast radiative transfer in participating media are discussed. Various numerical solution techniques are discussed and the major advantages and challenges to each are noted. Applications of ultrafast laser transport are discussed.

INTRODUCTION
In recent years, the advent of ultrashort-pulsed laser technology (pulse widths in the ps and fs range) has allowed for great advances in biomedicine, with applications including optical imaging [1,2], optical tomography [3], and laser irradiation/ablation of cancerous tumors from surrounding healthy tissues [4-6]. In such applications, radiation is a dominating mode of heat transfer, and it is critical that radiative processes are accurately predicted. With the emergence of high-powered computers, numerical modeling of radiative transfer has become an appealing and cost-effective alternative to full experimentation. In general, contributions from radiation can be determined via solution of the Equation of Radiative Transfer (ERT). For cases involving ultrafast transport, neglecting time dependence in the governing hyperbolic wave equation can lead to significant error, due to the short time-duration of the transport processes. To accurately determine ultrafast radiative transfer, solutions of the transient ERT (TERT) are required, the general vector form of which is given below:

\[
\frac{1}{c} \frac{\delta I(r, s, t)}{\delta t} + s \cdot \nabla I(r, s, t) = -\beta I(r, s, t) + \sigma_a I_a(r, t) + \frac{\sigma_t}{4\pi} \int I(r, s', t) \Phi(s', s)d\Omega',
\]

In the preceding equation, the terms on the left-hand side represent the temporal and spatial gradients of intensity, and the terms on the right-hand side represent intensity augmentation and attenuation due to absorption, scattering, and blackbody emission. When irradiation from ultrafast lasers is concerned, blackbody emission is generally negligible and radiation transfer is spectral.

In this work, we recall several numerical methods for...
solving the governing TERT and the advantages/limitations for each. The methods discussed in this treatise are: Monte Carlo Method (MCM), Discrete-Ordinates Method (DOM), Finite Volume Method (FVM), Diffusion Approximation (DA), Spherical Harmonics (Pₙ), Integral Method (IM), Discrete Transfer Method (DTM), and Radiation Element Method by Ray Emission Model (REM²). We also discuss practical applications of ultrafast radiative transfer and research contributions in the field, in some detail.

MONTE CARLO METHOD

Statistical methods, such as the Monte Carlo method, are commonly implemented to solve complicated physical problems due to their mathematical simplicity. For radiative transfer, accurate solution of the integro-differential TERT is a formidable task. The MCM involves the tracing of photon bundles from their point of emission to absorption [7], with direction changes experienced due to reflection, refraction, and scattering. Physical properties such as intensity and heat flux are calculated using statistical averages. These processes are simple but extremely time-consuming and subjected to statistical errors. The MCM is sometimes considered as an alternative to realistic experiments.

Jacques [8] determined time-resolved irradiation propagation in a turbid tissue due to both fs and ps laser pulses, investigating the impact of scattering albedo and medium anisotropy on the results. Later, Hasegawa et al. [9] investigated the interaction of near-infrared light with tissue slabs using the MCM in order to clarify the microscopic Beer-Lambert law. Works by Guo et al. [10, 11] used the MCM to directly determine transient radiative transport due to short-pulsed laser interactions in two- and three-dimensional participating media. Guo et al. [10] analyzed the effects of boundary conditions, pulse width, beam radii, and scattering anisotropies on transient results. Further, Guo et al. [11] compared MCM results with experimental measurements from a 60 ps pulsed laser transmission, finding that their statistical model accurately conformed to the experimental values.

More recently, Lu and Hsu [12, 13] introduced a reverse MCM for determining ultrafast radiative transfer, by which photon bundles are tracked in a time-reversal manner. This method is claimed to be more computationally efficient than the conventional MCM, due to the fact that it is not necessary to track photons that fail to reach a collector site during simulation. However, this method may be inappropriate if detailed physical information is required [12]. Comparisons with both the conventional MCM and DOM found a good overall agreement. The difference between MCM for ultrafast radiative transfer and MCM for traditional radiative transfer lies in the accounting of photon flight times.

DISCRETE-ORDINATES METHOD

One of the more common approximate methods for solving the TERT is the Sₙ discrete-ordinates method [14]. Using the DOM, the continuous angular variation is represented by a finite number of discrete radiation directions, and solid angle integrations are replaced by quadrature sums [7, 15]. Fiveland [15] and Truelove [16] were among the first to implement the DOM for determining radiative heat transfer, investigating radiative processes in three-dimensional enclosures containing participating media.

Mitra and Kumar [17] first investigated the solution of the TERT using the DOM for examining short light pulse transport through a 1-D medium, comparing DOM results with other solution methods. Mitra and Churnside [18] also investigated the solution of the TERT for oceanographic lidar using the DOM. In 2001, Guo and Kumar [19] first developed the transient DOM (TDOM) to analyze the interaction of short-pulsed laser light with 2-D turbid media, in which they found the DOM solutions to be in good overall agreement with MCM solutions. In 2001, Guo [20] further presented the first formulation of TDOM for solving ultrafast radiative transfer in 3-D participating media.

Guo and co-authors extended the study of the transient DOM to various ultrafast applications, including Duhamel’s superposition theorem for the treatment of pulse train irradiation [21, 22], formulation in cylindrical axisymmetric coordinates [23], and combination with heat conduction [4,5,24]. Additionally, Sakami et al. [25] combined the TDOM with piecewise parabolic interpolation to analyze the behavior of a scattering medium irradiated by a laser pulse.

More recently, Hunter and Guo [26] developed a new phase function normalization technique to accurately conserve both scattered energy and phase function asymmetry factor after DOM discretization, applying the technique to determine transient radiative transfer in a turbid media subject to uniform collimated irradiation [27]. They found that it is imperative that both quantities are conserved in order to accurately predict transient propagation of radiant energy. The DOM has generally been found to be accurate and computationally efficient [28,29], though it suffers a drawback due to ray effect (especially for optically thin media) and false scattering inherent from the directional approximation of the continuous angular variation.

FINITE VOLUME METHOD

The finite volume method was formulated for radiative transfer by Raithby and Chui [30] and expanded on by both Chui et al. [31] and Chai et al. [32]. This method is similar in nature to the DOM, but it does not involve the use of specific discrete directions. The ERT is directly integrated over both control volumes and solid angles, guaranteeing overall radiant energy conservation and reducing ray effect. In addition, the choice of directions and computational grid is extremely
flexible for the FVM, making the FVM a popular choice for more complicated analysis, such as irregular geometries. While previous FVM studies solely investigated steady-state radiative transfer, a series of publications by Chai [33,34] adopted the FVM for use in solving the TERT. Chai first applied the FVM to a 1-D slab subjected to both single-pulse and repeated-pulse collimated irradiation at a boundary, and compared results with those determined using the integral formulation [33]. He later expanded the formulation for use with 2-D irregular geometries [34] and, along with Hsu and Lam [35], modeled 3-D transient radiative transfer in a rectangular enclosure, once again finding good agreement with the integral formulation. Chai et al. investigated both the STEP and CLAM finite differencing schemes, noting that the CLAM scheme reduces errors and better represents steep radiative intensity gradients.

A more recent study by Muthukumar and Mishra [36] investigated Gaussian-profile short-pulsed laser interaction with an inhomogeneous planar medium made up of layers of varying optical thickness, in which they found the impact of optical depth and scattering albedo on the time spans of both transmittance and reflectance signals. Kim et al. [37] further investigated transient radiative transfer in a 1-D medium subject to radiative equilibrium, in order to simulate medium heating during irradiation via short-pulsed lasers (as in metals). Due to the full solid angle integration of the ERT, the FVM has been shown to be less computationally efficient than the DOM [28,29], although it has the advantage of flexibility in the choice of discrete directions. The DOM adopts a FVM strategy in spatial discretization, in order to take advantage of the geometric flexibility associated with the FVM.

DIFFUSION APPROXIMATION

The diffusion approximation [38] can be used to greatly simplify the ERT and has been widely used to describe photon migration and particle transport in diffusing media. The DA was first introduced as a means of determining photon transport in stellar bodies. Ishimaru [39] investigated the DA as a method of determining the diffusion of light in turbid media, noting that the approximation was valid for optically thick, weakly-forward scattering media.

Yoo et al. [40] compared the DA to experimental results, noting that time-dependent photon transport in a slab of random media is accurately predicted by the DA only when the medium is much thicker than the mean-free path. Slizard and Pomraning [41] compared the DA to direct ERT discretization for radiation penetrating a cold slab. Brewster and Yamada [42] determined time-dependent reflectance and transmittance in an optically thick turbid media subjected to pulsed irradiation using the DA, and compared their results to both the MCM and experimental values, noting that parameters determined using the DA are accurate enough to calculate medium optical properties. Further work by Furutsu and Yamada [43] determined that the diffusion coefficient should be independent of absorption, and introduced applications for the DA, such as optical tomography.

More recently, Contini et al. [44] investigated time-dependent propagation of light in both a turbid slab and semi-infinite medium using the DA, determining that it gives excellent descriptions of photon migration through a slab for varying optical properties. Guo et al. [45] compared DA results with the MCM for ultrafast laser transport in a 3-D enclosure containing inhomogeneities and/or a highly-absorbing layer. They determined that the DA is useful for describing light transport in highly scattering tissues, but that there are severe limitations in predicting ballistic photon transport at boundaries or with the presence of an absorbing region, making the DA less suitable than direct radiative transfer methods.

OTHER METHODS

The spherical harmonics, or PN method, is an approximate method that eases solution procedure complexity by transforming the integro-differential governing equation into a set of partial differential equations. The method was first introduced by Jeans [46], and was used by Menguc and Viskanta [47,48] to predict radiative transfer in 2/3-D enclosures. Kumar et al. [49] first developed the hyperbolic P1 approximation for the solution of the TERT, analyzing transmittance and intensity through a one-dimensional sample. They compared hyperbolic results with parabolic approximations, determining that the hyperbolic approximation was more accurate for thin media than the corresponding parabolic approximations. Yamada and Hasegawa [50] applied the P1 approximation to determine time-dependent photon migration in biological tissue, noting that errors manifested at small times when compared with MCM results. Mitra et al. [51] also used the P1 approximation to determine transient radiative transfer in a 2-D rectangular enclosure.

A review by Mitra and Kumar [52] solved the transient ERT using the P1 approximation, the two-flux method, and the discrete-ordinates method, and compared the ability of each approximation to accurately predict short-pulse light transport through a 1-D absorbing-scattering slab. They noted that different approximations produced varying speeds of propagation. Bhuvenswari and Wu [53] determined ultrafast radiative transfer in a planar media subject to pulse irradiation using the P1, P1/3, P1 parabolic, and DOM methods, finding that P1/3 was the most accurate when compared with the DOM. The P3 approximation has not widely been used in recent years, due to the improved accuracy and efficiency of other approximate solution methods.

The Discrete Transfer Method (DTM), developed by Lockwood and Shah [54], is an approximate method that holds many similarities to the DOM in nature. The DTM implements a ray-tracing procedure [7, 55], by which rays are sent out from boundary nodal points in pre-determined directions, and the ERT is then solved for each ray as it travels through the medium.
Rath et al. [55] first examined the solution of the TERT using the DTM, considering the impact of a short-pulsed laser on the boundary of a 1-D planar medium. They compared their results with those determined by Sakami et al. [25] using the DOM, and found good overall agreement. Mishra et al. [28] analyzed the impact of short-pulse laser irradiation on transmittance and reflectance signals in a 1-D planar medium using the transient DTM, and compared the results with DOM and FVM. They found that all three methods attain comparable solutions, but that the DOM and FVM are more computationally efficient than the DTM, due to the lack of a time-consuming ray tracing procedure in those methods.

Approximate differential methods are widely used to solve the TERT, due to their relative simplicity and ease of implementation, but they suffer from certain drawbacks, including difficulty in accurately capturing wave propagation and inherent errors due to ray effect, false scattering, numerical diffusion, etc. To this end, the integral method [56, 57] was developed, in which the ERT is directly integrated based on geometric constraints. This eliminates diffusion error and ray effect due to the approximation of a continuous angular variation using quadrature sets.

Wu and Wu [58] and Wu [59] first solved the TERT using the IM, applying it to both 1-D planar and 2-D cylindrical linearly anisotropic scattering media. They found that results determined using this method both highly accurate and efficient, and matched well to previously published MCM solutions. Tan and Hsu were also among the first to apply the IM to determine transient radiative transfer, investigating wave behavior and propagation in a 1-D slab [60], and then extending the method for a 3-D enclosure housing a participating medium [61]. In both treatises, they found that steady-state heat fluxes and intensities matched well with those determined using the MCM. While the IM is more ideal in nature, the difficulty of integration highly depends on medium geometry and properties, making it difficult to employ in real situations. However, it still remains one of the more accurate methods for determining transient radiative transfer.

The Radiation Element Method by Ray Emission Model (REM²), introduced by Maruyama [62] and Maruyama and Aihara [63] and expanded on by Guo and Maruyama [64], is a generalized zonal method, in which the governing ERT is solved integrally and the ray tracing method was employed to find radiant-energy fraction based view factors.

Guo and Kumar [65] first extended the REM² method for transient radiative transfer in a plane-parallel system, comparing results to the MCM for both diffuse and collimated boundary problems. They also investigated the sensitivity of radiation element size, ray emission number, and time increment, finding that the size of the time-step should be proportional to the radiation propagation time. Guo et al. [64] further extended the method for transient radiative transfer in a 3-D medium, finding good agreement when results were compared to the MCM.

**ULTRAFAST LASER APPLICATIONS**

**Optical Imaging and Optical Tomography**

The development of ultrafast laser technology has allowed for great advances in near-infrared optical imaging [1-3] of growths imbedded in biological tissue, such as cancerous tumors. Imaging of inhomogeneities with continuous-wave (CW) light sources results in the washing out of shadows by multiple random scattering of light, due to the highly scattering nature of tissue [67]. Ultrafast laser technology, coupled with sophisticated time-resolved imaging techniques, allows for the detection of optical information without detecting such multiple-scattered light caused by diffuse radiation. In a theoretical investigation of 3-D imaging properties under ultra-short-pulsed illumination, Gu and Sheppard [68] reported large increases in image resolution compared with CW laser illumination of similar wavelength. In cases where diffuse radiation dominates, such as highly turbid media, diffuse optical tomography [3, 69, 70] was proposed. Diffuse optical tomography involves inverse image reconstruction using experimental measurements and continuous forward modeling of photon migration with predetermined optical parameters. Guo et al. [71] introduced a simple imaging method for initial screening in which complicated, time-consuming inverse optimization is not needed.

Wang et al. [72] implemented an ultrafast optical shutter to image objects hidden behind various scattering walls, including human breast tissue, achieving submillimeter resolution. Zaccanti et al. [73] investigated picosecond pulse transmission through a thick turbid media, determining the potential for detecting absorbing structures using time-gated scanning imaging. Later, Alfano and co-authors [2, 67] implemented ultrafast time-gated optical detection to isolate the early-arriving portion of ultrafast laser pulses to locate both opaque and translucent inhomogeneities buried in thick biological tissue, in addition to investigating the nonlinear optical tomography of human organs [74], indicating that the use of ultrafast light for imaging biomedical media would allow for the development of inexpensive and noninvasive clinical imaging techniques for diagnostic purposes.

Villringer and Chance [75] implemented ultrafast laser technology for non-invasive optical spectroscopy of human brain function in vivo. Benaron et al. [76] also investigated applications for the human brain, analyzing photon transit time for low-power light passing through the skull and brain for tomographic imaging of hemoglobin oxygenation. Wu et al. [77] performed fluorescence tomography, using a picosecond laser pulse with a single photon to detect objects in thick, turbid media, indicating the potential for this technique to detect breast tumors. Schmidt et al. [78] constructed a multichannel, time-resolved imaging device to detect locations of small inhomogeneities in medical optical tomography. Ntziachristos et al. [69] found that diffuse optical tomography of human breast tissue in vivo provides for localization and quantification for tissue chromophore concentrations, and provides images
that are accurate compared to magnetic resonance imaging. Tromberg et al. [79] and Hebden et al. [80] additionally investigated optical tomography with picosecond laser pulse for imaging in human breast tissue.

Later, Quan and Guo [81] developed a fast 3-D optical imaging method utilizing an exogenous fluorescence agent, by which ultra-short laser pulses illuminate tissue and excite fluorescence, allowing for 3-D image reconstruction of relative fluorescence distribution. They showed the ability of this method to accurately image a small tumor in turbid media. In a related work, Wan et al. [82] developed an image-projection approach based on slope analysis of time-resolved back-scattered signals which was able to detect the presence, size, and location of an inhomogeneity in turbid tissue. Guo et al. [71] additionally developed a log-slope difference mapping approach for detection of cancerous tumors in human breast tissue using near-infrared ultrashort laser pulses.

**Laser Biomedical Processing**

In addition to optical tomography, ultrashort lasers have become widely used for various laser biomedical processing applications, which include precision laser microsurgery [83-85], laser-tissue welding [23, 86-88], and tissue microprocessing. A popular biomedical application for ultrashort lasers is precise material removal and micromachining via plasma-mediated ablation [6, 89-100]. During plasma-mediated ablation via ultrashort lasers, the timescale of plasma formation due to laser material interaction is much shorter than the material thermal relaxation time, which limits/eliminates thermal diffusion [90,91]. This results in an effective minimization of thermal and mechanical damage that can occur during ablation with CW lasers [92,93], an important result for interaction with biological tissue in vivo.

An early study by Niemz et al. [92] investigated plasma-mediated ablation on human donor corneas, measuring the mean rate of tissue removal and finding that laser excisions were smooth with little distortion. They further indicated that use of a mode locked Nd:YLF laser would be a reasonable alternative to excimer lasers for corneal surgery. Another work by Kautek et al. [93] performed femtosecond-pulse laser ablation of corneal tissue, finding that ablations were also of high quality. Fischer et al. [94] and Loesel et al. [95] investigated picosecond-pulse ablation of brain tissue, and found that no thermal damage was induced. Oravecky et al. [96] investigated plasma-mediated ablation with femtosecond pulses, finding that ablation craters were obtained with no thermal diffusion to the surrounding, and demonstrating the ability of ultrafast lasers to act as precise microsurgical tools. Works by Feit et al. [97] and Kim et al. [98] presented, in detail, the physical mechanisms associated with plasma-mediated ablation and an investigation into ablation characteristics, such as differences in ablation crater morphology with varying pulse-width.

Huang and Guo [90,91] used ultra-short pulsed lasers to achieve thin-layer separation of both in vitro wet [90] and freeze-dried human dermal tissues [91], finding no thermal damage to occur with single line ablation and insignificant thermal damage with multiline ablation in both studies. In addition, further works by Guo and co-authors [99,100] demonstrated the ability to remove thin layers of biofilm contamination via picosecond laser ablation, removing contaminating layers of blood from various substrates neatly and without experiencing thermal diffusion damage. A work by Jiao and Guo [6] modeled ultrashort pulsed laser ablation in both water and biological tissue, in order to accurately predict laser intensity distribution in turbid tissue without costly experimentation, finding excellent agreement with experimental results.

Additionally, use of the thermal mechanism of ultrafast lasers was also studied recently. Jaunich et al. [4] investigated subsurface ablation on in vitro, freshly excised mouse tissue, and compared experimental results with a developed numerical model that combined the bio-heat transfer equation with a hyperbolic conduction model, finding good agreement. A numerical study by Jiao and Guo [5] looked at the thermal response of tissue from an ultrashort focused laser beam, finding that focused beams were able to produce higher temperature rise at the target area than in surrounding healthy tissue, making it an ideal method for combating cancerous cells.

**Micromachining of Metals and Polymers.**

In addition to laser tissue processing and optical imaging, ultrafast lasers are commonly implemented for precise micromachining of various materials, including polymers and metals, via plasma-mediated ablation [101,102]. Distinct advantages of laser micromachining include the ability to drill and cut with high precision in situations where dimensional tolerance is extremely strict, and the ability to accurately machine submicron feature sizes for applications such as micro- and nano-electronics [102] or micro-patterning of biodegradable polymers to assist in drug delivery [103,104].

Corkum et al. [101] presented an early review of the thermal response of metals to excitation by ultrashort-pulsed lasers, developing the theory to describe optical damage’s dependence on pulse-duration, and validating their model by investigating optical damage to both molybdenum and copper experimentally. Liu et al. [102] presented a discussion of the mechanisms of ultra-short pulse laser ablation, describing in detail the differences between short- and long-pulse ablation. Precision micromachining results were presented, and they found that femtosecond pulses produced clean, uniform holes and cuts when used to cut into a polyethylene film. When long-pulses were used, they found uneven material cutting and cracking to occur. Femtosecond lasers were also shown to produce high quality drilled holes through steel.

Kanchlarla and Chen [103] used direct laser writing and percussion drilling with ultraviolet lasers as a means of micropatterning and fabricating biodegradable micro-devices.
out of both poly-D-lactic acid and polymer poly-vinyl alcohol. They witnessed minimal thermal damage when producing said devices. Aguliar et al. [104] also investigated micro patterning of biodegradable polymers using femtosecond lasers, performing in vitro degradation tests of the polymeric material. They found that their laser-patterned samples were within one standard deviation of control samples, concluding that ultrafast lasers are beneficial tools for micro-patterning biodegradable polymers. Both Kancharla and Chen [103] and Aguliar et al. [104] stressed the importance for micromachining of biodegradable polymers with regards to biomedical applications, such as precision drug delivery.

Zergioli et al. [105] investigated the micro deposition of metals using femtosecond laser pulses, achieving high-quality chromium and indium oxide microdeposition on glass and silica and performing serial writing of both dots and lines with submicron resolution. Chimmalgi et al. [106] and Hwang et al. [107] further explored micromachining with femtosecond lasers, performing controllable surface nanomachining on thin gold films [106] and fabricating both straight and 3-D micro-channels with good wall-surface quality on glass [107]. Later, Crawford et al. [108] investigated groove micromachining in silicon with femtosecond pulses, determining the impact of ablation rate, groove depth, and number of ablation passes on overall groove quality.

Srisungsithisunti et al. [109] extended the application of ultrafast lasers in micromachining to microimaging applications, using direct femtosecond laser writing to fabricate volume Fresnel zone plates inside fused silica. Volume Fresnel zone plates are designed to focus light together, which yields a higher diffraction efficiency for microimaging. Chowdhury and Xu [110] numerically predicted heating, melting, and evaporation of metal under irradiation from femtosecond lasers, finding that a one-dimensional, parabolic, two-step model is able to achieve accurate agreement with experimental data. In another numerical study, Cheng and Xu [111] investigated the mechanisms behind nickel decomposition during laser ablation through use of molecular dynamics. They witnessed two distinct laser fluence regimes caused by different ablation mechanisms. For low laser fluence, material decomposition occurs due to phase explosion, while for high laser fluence, critical phase point separation occurs.

The applications of ultrafast lasers listed here, for both biomedical, imaging, and material processing applications, are not exhaustive. However, the extensive use of ultrafast lasers for many applications in different fields indicates the importance and power of this technology. The authors acknowledge that there are many excellent works that may not have been properly cited here.

CONCLUDING REMARKS
Various advances in modeling ultrafast radiative transport in participating media, as well as advances in ultrafast laser applications, have been discussed in this work. It is important to note that the publications, methods, and applications cited in this treatise are not exhaustive (there are many more not listed here). The advances in modeling ultrafast radiative transfer have progressed greatly in recent years, leading to more accurate description of radiative processes without the necessity of full experimentation.

REFERENCES


