Invited Paper

Physical mechanisms of pulsed infrared laser ablation of biological tissues

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ABSTRACT

Time-resolved measurement of the stress transients generated by pulsed infrared irradiation and ablation of tissue has demonstrated that these processes are governed primarily by photothermal processes. For ablation of porcine dermis at 2.79 μm (Q-sw Er:YSGG) and 10.6 μm (CO_2), the onset of material removal has been shown to be delayed with respect to irradiation and the magnitude of the generated stress transients are consistent with a model for explosive material removal. Upon consideration of the threshold radiant exposure for ablation, it appears that the mechanism and dynamics of these processes are controlled by explosive boiling as the tissue water is likely to be significantly superheated.

To examine this issue further, we employed time-resolved optical interferometry to measure the surface displacement generated by Q-sw Er:YSGG laser irradiation of pure water for radiant exposures below the ablation threshold. This was done to directly measure the dynamic thermal expansion and interphase mass transfer generated by pulsed laser heating. These results are compared to a model which computes the dynamic thermal field within a semi-infinite pool of water undergoing pulsed irradiation while subject to a surface heat flux condition given by kinetic theory. We find that the measured mass fluxes exceed that predicted by simple kinetic theory arguments. The implications of the experimental and model results to pulsed laser ablation of tissue are discussed.

Keywords: laser ablation, explosive boiling, tissue ablation, interphase mass transfer

1. INTRODUCTION

Pulsed infrared lasers have found extensive use for medical applications due to their ability to hemostatically incise and excise tissue. In particular, pulsed CO_2 and erbium lasers are receiving much attention for burn wound debridement and aesthetic skin surgery. The ability of infrared lasers to achieve these effects is due to the significant optical absorption of water at several infrared wavelength bands and the high water content (65–90%) of soft biological tissues. The tissue effect achieved by pulsed infrared lasers do not extend to surgical lasers operating at other wavelengths. For example an ultraviolet laser, emitting radiation with an optical absorption coefficient in tissue identical to an infrared laser wavelength, usually cuts tissue with greater efficiency but is unable to do so hemostatically. This difference in tissue effect is likely due to the optical absorption of ultraviolet radiation by tissue proteins rather than by tissue water.

There is some evidence that the different tissue effect produced by radiation absorbed by structural proteins results from a difference in the ablation mechanism at a fundamental thermodynamic level. Studies which measure the mechanical recoil resulting from ablation using radiation absorbed by structural proteins demonstrate that ablation proceeds via a rapid surface vaporization process.[1–3] That is, for ultraviolet radiation, the recoil stresses produced by tissue ablation can be quantitatively modeled as a laser driven gas-dynamic process whereby the tissue

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is transformed from a condensed phase to a vapor phase. By contrast, studies which measure the ablative recoil generated by radiation absorbed primarily by tissue water do not support a rapid surface vaporization model. Instead it appears that such processes are explosive in nature.[4]

In this paper, we review evidence which indicates that, when water is the primary chromophore, pulsed laser ablation is driven by superheating of the tissue water leading to explosive boiling. Then, to characterize the thermal events which may be responsible for such a process, we consider a model for non-equilibrium interphase heat and mass transfer resulting from pulsed infrared laser irradiation of water. This model predicts the mass transfer and temperature distribution within the liquid bulk using a surface boundary condition given by kinetic theory. We compare the model results with measurements of mass removal generated by pulsed Er:YSGG (λ=2.79 μm) laser irradiation of a water pool using a time-resolved interferometric method. The implications of the model and experimental results are discussed.

2. THE EXPLOSIVE NATURE OF PULSED TISSUE ABLATION

The explosive nature of laser ablation processes driven by the optical absorption of tissue water was first verified by a study of plume dynamics of Er:YAG laser ablation of skin and bone.[5] This study demonstrated that there is a significant temporal delay between the end of laser irradiation and the onset of material removal. Further, the propagation velocities of the ablation plume were roughly consistent with models of shock wave dynamics generated by a focal explosion.

Subsequently, stress transients generated by pulsed Er:YSGG and TEA CO_2 laser radiation of skin were measured quantitatively.[4] These measurements were consistent with the observation made in the earlier plume dynamics study that the onset of material removal is delayed relative to laser irradiation. In addition, the magnitude of the measured stress transients were inconsistent with gas dynamic models for rapid surface vaporization that were shown to be applicable for processes where tissue proteins are the primary chromophore.[2] Instead, the measured stress transients showed excellent agreement with a model for explosive material removal initially developed for pulsed laser ablation of polymers and metals.[6] Moreover, when one assumes that heating occurs under isobaric, adiabatic conditions, the temperature of the tissue water is raised to $\sim$260–280°C at the threshold energy dose for material removal. For atmospheric pressure, these temperatures are comparable to both the spinodal limit (305°C)[7] and the highest superheat temperature experimentally achieved in water (279.5°C).[8] This indicates that explosive boiling or phase explosion should be considered as a candidate mechanism for laser ablation of tissue in cases where water is the dominant chromophore.

To examine the temperatures achieved within water for a rapidly heated tissue, we constructed a model to predict the temperature distribution resulting from pulsed laser irradiation of a water sample. This model predicted the interphase heat and mass transfer rates using kinetic theory to provide upper bounds on this non-equilibrium transport process. To verify the model predictions we determined the mass transfer rates by measuring the surface displacement of a water pool exposed to pulsed Q-sw Er:YSGG laser irradiation.

3. INTERPHASE HEAT AND MASS TRANSFER RESULTING FROM PULSED LASER IRRADIATION

3.1. Theory

The absorption of pulsed laser radiation by a sample produces a discontinuity in thermodynamic properties at the interface between the sample and its surroundings. When the discontinuity is large in magnitude, the resulting heat and mass transfer are high rate processes poorly described by standard diffusion equations (i.e., Fick's and Fourier's law). However, using kinetic theory, it is possible to derive upper bounds for the heat and mass fluxes that are generated when a condensed sample in contact with a vapor phase is suddenly heated.

Consider a water system at uniform temperature T in equilibrium with its vapor. The vapor pressure above the condensed sample is the saturation pressure $p_{sat}(T)$. Assuming ideal gas behavior and that all vapor molecules which collide with the surface of the condensed phase get assimilated, kinetic theory states that the flux of vapor molecules absorbed by the condensed phase $j_{v\to c}$ is:

$$j_{v\to c} = n_v \sqrt{\frac{k_B T}{2\pi m}}, \tag{1}$$

where n_v is the number density of vapor molecules, k_B is the Boltzmann constant and m the molecular mass. To preserve equilibrium this 'condensation' flux is balanced by a flux of molecules which are spontaneously emitted as vapor. If we consider a situation where a vacuum is maintained above the condensed material, the condensed phase will spontaneously evaporate at the rate

$$j_{c \to v} = n_v \sqrt{\frac{k_B T}{2\pi m}}. \tag{2}$$

This upper limit for spontaneous emission from a condensed phase allows us to place an upper bound on the heat flux q_{max} generated by such an interphase mass transfer process

$$q_{max} = \rho_v h_{fg} \sqrt{\frac{k_B T}{2\pi m}}, \tag{3}$$

where h_{fg} is the latent heat of vaporization and ρ_v is the mass density of the vapor.

With these general results we can address the situation where a short, energetic laser pulse irradiates a water sample initially at equilibrium with saturated atmospheric air. The vapor pressure in the air will be the saturation vapor pressure, approximately 2300 Pa. The absorption of laser radiation results in a temperature rise in the water and a spontaneous vapor emission rate greatly exceeding the rate at which water vapor molecules collide with and are assimilated by the liquid surface. Thus non-equilibrium conditions are established locally at the liquid-air interface resulting in high-rate interphase heat and mass transfer. Eqs. (2) and (3) assume that there is no 'condensation' flux and this should provide upper bounds for the net interphase mass and heat fluxes. In reality, such high fluxes will be sustained for only a short time due to the presence of the atmosphere. The surrounding molecules will tend to redirect the water vapor molecules towards the liquid surface serving to re-establish local equilibrium in short order. Once this occurs, conventional diffusive and convective models for heat and mass transfer are adequate to describe the subsequent evolution of the system. However, if the laser pulse is sufficiently energetic that it raises the water temperature above 100°C, the saturation vapor pressure will exceed atmospheric pressure and result in the emission of a shock wave. The emitted water vapor will displace the surrounding atmosphere and eqs. (2) and (3) will provide reasonable approximations for the net interphase mass and heat fluxes for a longer time interval.

3.2. Modeling

The interphase mass and heat fluxes have a significant effect on the temperature distribution within the irradiated sample. The temperature distribution within a condensed medium undergoing high-rate interphase heat transfer is determined as follows. We consider the problem in one spatial dimension which implicitly assumes the transverse width of the laser beam to be much larger than the optical penetration depth of the radiation. The governing thermal diffusion equation is

$$\frac{\partial^2 T(z,t)}{\partial z^2} = \frac{1}{\alpha} \frac{\partial T(z,t)}{\partial t} - \frac{q'''(z,t)}{k}, \tag{4}$$

where T is temperature, z the axial position, t time, α the thermal diffusivity of the condensed phase, q''' the volumetric energy density supplied by the laser and k the thermal conductivity of the condensed phase. The initial condition is that the condensed phase is at ambient temperature T_∞. The boundary condition at the surface is specified by the interphase energy flux i.e.,

$$k \left. \frac{\partial T}{\partial z} \right|_{z=0} = \rho_v h_{fg} \sqrt{\frac{k_B T(z=0,t)}{2\pi m}}. \tag{5}$$

This system of equations was solved using the finite element method in z and Runge-Kutta numerical integration in t. The time history of the temperature field for pure water is shown for a radiant exposure of 1000 J/m^2 in Fig. 1, for an absorption coefficient $\mu_a = 5.15 \times 10^5$ m^{-1}, a laser pulse duration (FWHM) of 50 ns and an ambient temperature T_∞=293 K.

The numerical solution for the temperature field can be used to predict the surface displacement Δz of a liquid sample subject to high-rate interphase heat and mass transfer. The surface displacement is a function of (a) the thermal expansion of the liquid, (b) the material vaporized from the liquid surface and (c) the compression of the

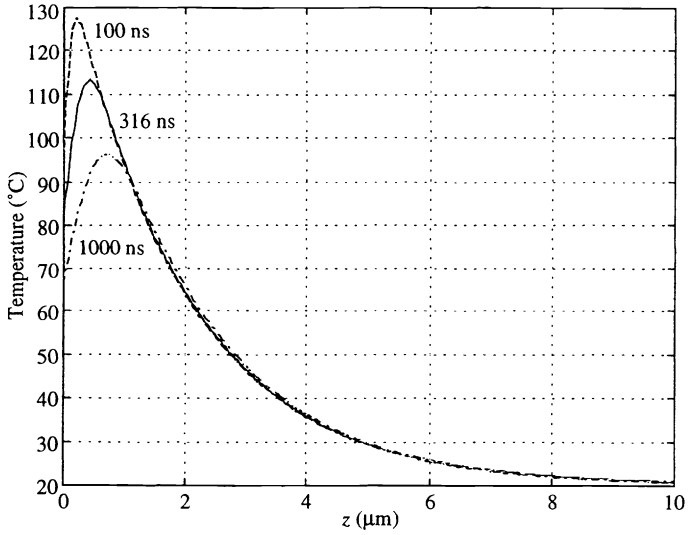

Figure 1. Predicted temperature field at $t = 100$, 316 and 1000 ns in pure water when exposed to a laser pulse with $t_p = 50$ ns, $\varepsilon_0'' = 1000$ J/m^2, $\mu_a = 5.15 \times 10^5$ m^{-1}.

condensed liquid due to the recoil force exerted by vapor molecules leaving the surface. Thus, the surface displacement may be expressed as the sum of these three contributions i.e.,

$$\Delta z(t) = \int_0^\infty \left[\frac{\rho_c(T_\infty)}{\rho_c(T(z,t))} - 1 \right] dz - \int_0^t \frac{q_{max}}{\rho_c h_{fg}} dt + \int_0^\infty \epsilon_r(z,t)\, dz, \tag{6}$$

where ρ_c is the density of the condensed liquid and ϵ_r the strain generated by the recoil of the vapor molecules leaving the liquid surface.

We can immediately evaluate the first two terms in eq. (6) as $\rho_c(T)$ is given by the thermophysical properties of water, $T(z,t)$ is given by the solution to the one dimensional heat transfer problem specified by eqs. (4) and (5) and the maximum interphase heat flux is given by eq. (3). To evaluate the recoil stress $\epsilon_r(z,t)$ we assume linear elastic behavior in the liquid and a hydrostatic state of stress. Thus the recoil strain in the z direction is proportional to the recoil stress, σ_r, via the bulk modulus, K_b, i.e.,

$$\epsilon_r(z,t) = \frac{\sigma_r(z,t)}{K_b}. \tag{7}$$

The recoil stress propagates into the liquid at the sonic velocity c_a and is equal to the product of the mass flux and the average velocity of the molecules leaving the liquid surface:

$$\sigma_r(z,t) = \sigma_r\left(z = 0, t - \frac{z}{c_a}\right) = m j_{c \to v} \frac{\bar{c}}{4}, \tag{8}$$

where $\bar{c}/4$ is the average velocity of the vapor molecules moving away from the surface. Assuming a Maxwellian velocity distribution, this is given as $(\bar{c}/4) = \sqrt{k_B T(z=0)/2\pi m}$.

$$\epsilon_r(z,t) = -\frac{\rho_v k_B}{2\pi m K_b} T\left(z = 0, t = t - \frac{z}{c_a}\right). \tag{9}$$

With eq. (9) giving the recoil strain we can evaluate the third term in eq. (6) and predict the surface displacement Δz.

3.3. Experiment

To determine the significance and extent of interphase heat and mass transfer during laser irradiation, we measured the surface displacement of pure water samples irradiated by the emission of a Q-sw Er:YSGG laser radiation with a modified Mach-Zehnder interferometer. A complete description of the interferometer instrumentation has appeared elsewhere.[9] This system is capable of measuring the displacement of a single location on the surface of a specularly reflecting sample with spatial and temporal resolutions of 1 nm and 4 ns, respectively. These capabilities make it an ideal tool to measure interphase mass fluxes.

The Q-sw Er:YSGG laser emits radiation at $\lambda = 2.79\,\mu$m with a duration of $t_p = 50$ ns (FWHM) and a maximum pulse energy of 25 mJ. The Er:YSGG laser beam irradiated the water sample at a slight angle ($\sim$ 12°) with a circular area 4 mm in diameter. IR quartz attenuators were used to adjust the laser pulse energy. As indicated in Figure 2, the HeNe laser probe beam of the interferometer intercepts the sample perpendicular to the water surface, has a diameter of approximately 15 μm at the air-water interface and is centered with respect to the Er:YSGG laser irradiation area.

The optical absorption of 2.79 μm radiation by water is $\mu_a = 5.15 \times 10^5\,\mathrm{m}^{-1}$ which corresponds to an optical penetration depth of 1.94 μm. Under these conditions, thermal diffusion during the 50 ns irradiation can be neglected as the characteristic thermal diffusion time across the optical penetration depth is approximately 3 μs. Also, all motion resulting from the generation of thermal stresses occurs during irradiation. This is because the characteristic time for a disturbance traveling at the sonic velocity c_a to traverse the optical penetration depth is given by $(\mu_a c_a)^{-1}$ and amounts to less than 2 ns; much less than the 50 ns irradiation time. Note that water vapor emitted during the laser pulse will not block the incident laser radiation since the absorption properties of water vapor in the IR are much weaker than they are in the condensed state. Moreover, even if water vapor absorbed the laser energy as strongly as condensed water does on a mass basis, the 10 nm of condensed water blown off during the pulse is negligible compared to the optical penetration depth.

Typical data traces are presented in Figure 3. As expected, the initial upward displacement is complete by 100 ns and corresponds to the timescale of energy deposition. The degree of downward surface motion depends on the maximum temperature attained within the heated sample which scales with the initial surface expansion. Almost no downward surface motion occurs when the pulsed energy deposition results in an initial expansion of less than 40 nm, while the downward surface motion which occurs after an initial upward motion of more than 100 nm is quite pronounced. The transition between the two cases occurs at an initial surface expansion of 60 nm and corresponds to an approximate initial surface temperature of 100°C. This agrees with the ideas put forth earlier, where it was argued that high rate interphase heat and mass transfer can only be sustained for an extended period of time when the surface temperature is raised above 100°C, so that the vapor emission can displace the surrounding atmosphere.

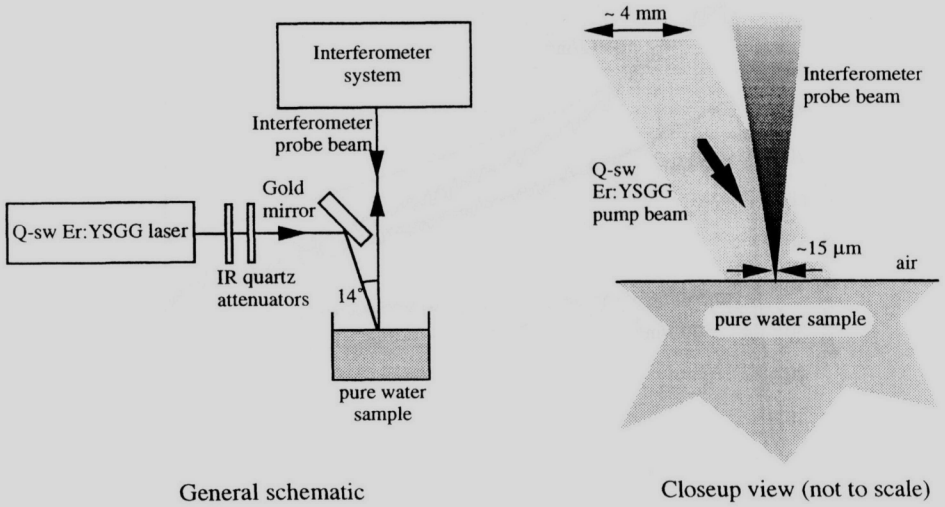

Figure 2. Experimental set-up for interferometric measurement of the surface displacement of a water sample.

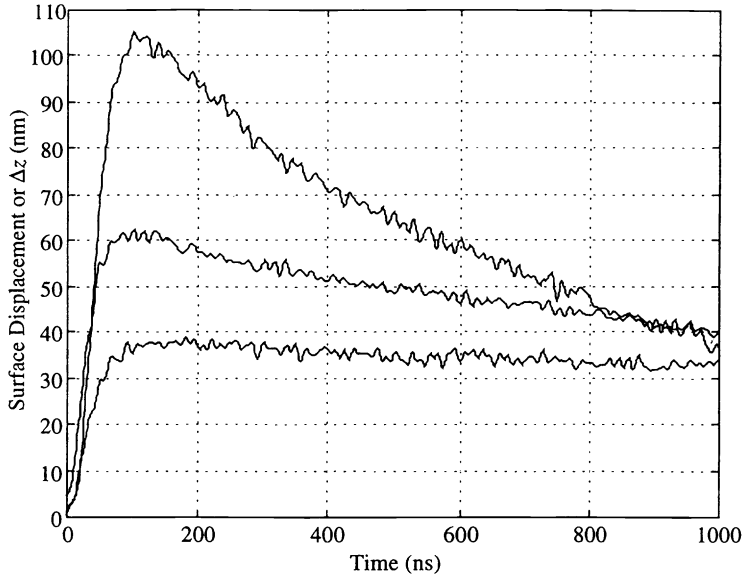

Figure 3. Typical surface displacement traces measured by the interferometer during Q-sw Er:YSGG laser irradiation of pure water.

4. DISCUSSION

The data traces were fitted to the model by matching the initial surface expansion. A comparison between typical data and the corresponding model result is depicted in Fig. 4. The radiant exposures indicated in the plot are the values used in the model to fit the measured initial surface expansion. As the figure clearly shows, for smaller initial surface displacements (<~60 nm), the model tends to overpredict the downward surface motion of the data. For larger initial surface displacements (>100 nm) the model tends to underpredict the downward surface motion. For intermediate initial surface displacements (~80 nm), there is good agreement between the model and the data. The maximum surface temperatures predicted by the model are 105°C, 97°C, and 70°C for the 1060 J/m^2, 890 J/m^2, and 500 J/m^2 traces respectively. One can use these surface temperatures in eq. (3) to determine the maximum predicted surface heat flux.

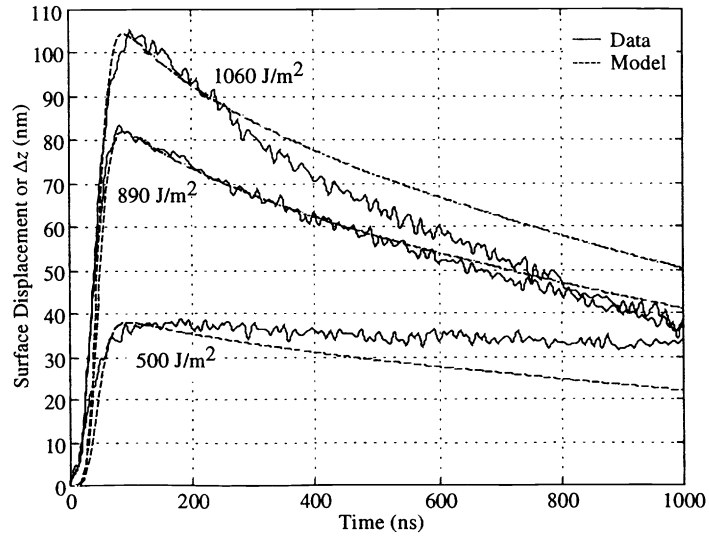

Figure 4. Comparison between data and model for Q-sw Er:YSGG laser irradiation of pure water.

The fact that the high-rate interphase heat and mass transfer model overpredicts the downward surface motion for small initial surface displacements is not surprising. Small initial surface displacements correspond to lower initial surface temperatures which results in spontaneous vapor emission rates insufficient to displace the surrounding atmosphere. However, what is surprising is that the high-rate interphase heat and mass transfer model *underpredicts* the surface displacement measured at the highest radiant exposure tested ($\varepsilon_0'' = 1060\,\mathrm{J/m^2}$) and results in a maximum surface temperature of approximately 105°C. The implication is that vapor leaves the surface at a rate which exceeds the upper limit predicted by kinetic theory.

Considering that, to our knowledge, no researchers have experimentally verified the limiting rates for interphase heat and mass transfer, it is possible that the simple estimates given by kinetic theory do not adequately capture the physics of the underlying physical phenomena. One possible explanation for the discrepancy is that the vapor was modeled as an ideal monatomic gas even though water molecules have significant rotational and vibrational energy modes. These extra modes may allow for higher non-equilibrium interphase heat or mass fluxes. Also, we assume that the velocity distributions of the molecules obey the canonical Maxwell-Boltzmann distribution, but in fact this may not be the case. The findings presented here must be independently corroborated and the associated theory may need to be reassessed.

5. IMPLICATIONS FOR ABLATION OF BIOLOGICAL TISSUES

The results shown have significant implications for laser ablation of biological tissues. Soft biological tissue is primarily water and its behavior in response to pulsed laser irradiation can often be modeled by that of water. Many researchers have pointed out that the ablation dynamics for certain combinations of wavelength, pulse duration, and tissue type are consistent with explosive boiling[4,10–12] which may result from a subsurface temperature maximum. Moreover, many researchers have found that the threshold radiant exposure for ablation using certain combinations of wavelength, pulse duration, and tissue type corresponds to surface temperatures well above 100 °C, if the tissue surface is assumed to be adiabatic.[4,12] It is clear that high-rate interphase heat and mass transfer will serve to quench the temperature maximum at the surface, producing a subsurface temperature maximum.

Figures 1 and 4 were generated using conditions similar to those existing during Q-sw Er:YSGG laser irradiation of water. In response to a 1000 $\mathrm{J/m^2}$ laser pulse incident on water, over 20°C of superheat is produced 200 nm below the surface about 300 ns after the laser pulse. Material removal driven by Q-sw Er:YSGG laser irradiation of tissue may occur when the absorption of energy produces enough subsurface superheat to nucleate a bubble that intercepts the tissue surface and spews out it contents. The time delay between the start of the laser pulse and the onset of ablation has been estimated to be on the order of a few hundred nanoseconds,[4,5,12] which would correspond to the time required for heat and mass transfer at the surface to establish a significant subsurface temperature peak. At this time, such a mechanism for Q-sw Er:YSGG laser ablation of water-based tissue is purely speculative. Until better measurements of the interphase heat and mass transfer are performed, no conclusions can be drawn as to the mechanism controlling the ablation threshold.

Previous studies have advanced thermal models for ablation which predict the existence of a subsurface temperature maximum that implies subsurface vapor nucleation and explosive boiling. Dabby and Paek developed a simple model for the temperature field of a target exposed to an intense laser which assumed that the target surface temperature could not exceed the target substance vaporization temperature.[13] This boundary condition is equivalent to forcing the interphase heat and mass flux to be zero for surface temperatures below the vaporization temperature and a varying heat and mass flux to clamp the surface temperature at the vaporization temperature for high rates of energy deposition. However, this model was criticized for its simplistic and non-physical treatment of interphase heat and mass transfer.[14,15] Miotello and Kelly have advanced a model for the temperature field in a laser irradiated target which includes a kinetic theory treatment of the interphase heat and mass transfer similar to that presented here.[14,15] They found that the subsurface temperature maximum resulting from surface cooling was not significant and likely did not influence laser ablation of aluminum. Our results differ from theirs only inasmuch as the thermophysical properties of water differ from that of aluminum.

More recent work has advanced the notion that a subsurface temperature maximum is not necessary for the production of subsurface bubble nucleation and explosive material removal.[4,14,15] If the duration of the laser pulse is sufficiently short and energetic, the water in the tissue target may be significantly superheated and driven in the vicinity of the spinodal temperature (305°C at 1 atm) at which point liquid water is intrinsically unstable leading to

homogeneous nucleation and thermal explosion. However, it must be noted that even in an ultrapure system where liquid water was heated without contact with solid boundaries, a superheat temperature of 279.5°C is the highest attained to date.[8] It is possible that in biological tissues the presence of dissolved gas and the multiple interfaces between the tissue water and structural proteins serve to catalyze the explosive boiling process at lower superheat temperatures.

6. CONCLUSION

Interphase heat and mass transfer may have an impact on the dynamics of laser ablation of biological tissue. For short, energetic laser pulses, a local non-equilibrium in both the temperature and water vapor concentration fields may lead to high-rate interphase heat and mass transfer where the net vaporization rate approaches the theoretical maximum predicted by kinetic theory. The choice of chromophore, whether it is the tissue water or the structural proteins, may determine the kinetics of the transport phenomena.

Numerical simulations of the surface displacement were computed for pulsed laser irradiation of pure water assuming that vapor emission proceeds at the maximum rate allowed by kinetic theory. When compared to these simulations, the data suggests that at high radiant exposures, interphase heat and mass transfer occurs at rates matching and exceeding the limits predicted by kinetic theory during Q-sw Er:YSGG laser irradiation of water. Independent confirmation of such high rates of interphase heat and mass transfer is clearly necessary.

The simple kinetic theory model described here may not capture the full complexity of the physics governing pulsed laser driven heat and mass transfer. More sophisticated models and numerical simulations are necessary. As the computational power available for numerical simulations increases due to technological advances, simulations of molecular dynamics could provide insight into how interphase heat and mass transfer proceeds for a system experiencing extreme local non-equilibrium. Such simulations could account for internal degrees of freedom as well as molecular systems whose velocity profiles undergo severe departures from the Maxwell-Boltzmann velocity distributions, as may be the case during intense pulsed laser irradiation. Further numerical simulation and experimental measurement should enable a full characterization of the events leading to the onset of material removal in pulsed ablation processes.

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