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Laser-ablation processes

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ABSTRACT

The physical mechanisms associated with ablation of matter by laser irradiation are quite different in different regions of parameter space. The important parameters are the laser wavelength; the laser flux versus time, position, and angle of incidence at the target; and the target properties as well as the properties of the laser-transport medium adjacent to the irradiated target surface. Important target properties include surface contour, laser reflectivity and absorption depth, thermal diffusively, vaporization energy, Grünelsen coefficient, spall strength, ionization energies and plasma opacity versus temperature and density. As the flux increases, the process becomes less dependent on most of these target properties. Depending on the values of these various parameters, at relatively low fluxes targets can be vaporized and these vapors can be transparent to the laser beam. If a transparent liquid or solid transport medium exists in front of the vaporized target material, then a complicated contained-vaporization process takes place and the work done on the target by the vapors can be several orders of magnitude larger than with a gas or vacuum transport medium; the degree of work enhancement can depend strongly on the vapor condensability and condensed matter thermal conductivity. For short-pulselength irradiations of semi-transparent targets with a low-acoustic-impedence-laser-transport medium adjacent to the target, ablation can occur by front-surface spallation. Above a certain flux level, the transport medium needs to be a vacuum in order for the beam to be able to propagate to the target. For targets in a vacuum exposed to fluxes of this order (and considerably higher) and for long pulselengths, most of the laser energy will be absorbed (before reaching the critical surface) by inverse bremsstrahlung in material blown off from the target; at higher fluxes, the beam will be stopped at the critical surface producing localized absorption along with much higher energy densities and non-thermal equilibrium behavior. When the combination of pulselength, beam diameter, flux and target material are such that the blowoff becomes opaque to the laser and also the blowoff can traverse many beam diameters during the pulselength, then a complicated radiation-hydrodynamic process is involved with strong feedback between blowoff hydrodynamic expansion, laser absorption, radiation transport, and target ablation by plasma reradiation. In this paper the various ablation processes and potential applications are reviewed from the threshold for ablation up to fluxes of about 10^{13} W/cm^2, with emphasis on three particular processes; namely, front-surface spallation, two-dimensional blowoff, and contained vaporization.

1. INTRODUCTION

The various mechanisms by which ablation of materials can be induced with lasers are discussed in this paper. The phrase laser ablation will be used to mean the removal of material by any physical process associated with laser irradiation. The important parameters associated with the laser irradiation conditions will first be discussed followed by a general description of the various processes of ablation and laser-material-interaction dependencies. Potential applications of three particular ablation processes will be discussed.

2. LASER-IRRADIATION CONDITIONS

The general conditions for irradiation of materials and important features are illustrated in Fig. 1. It is

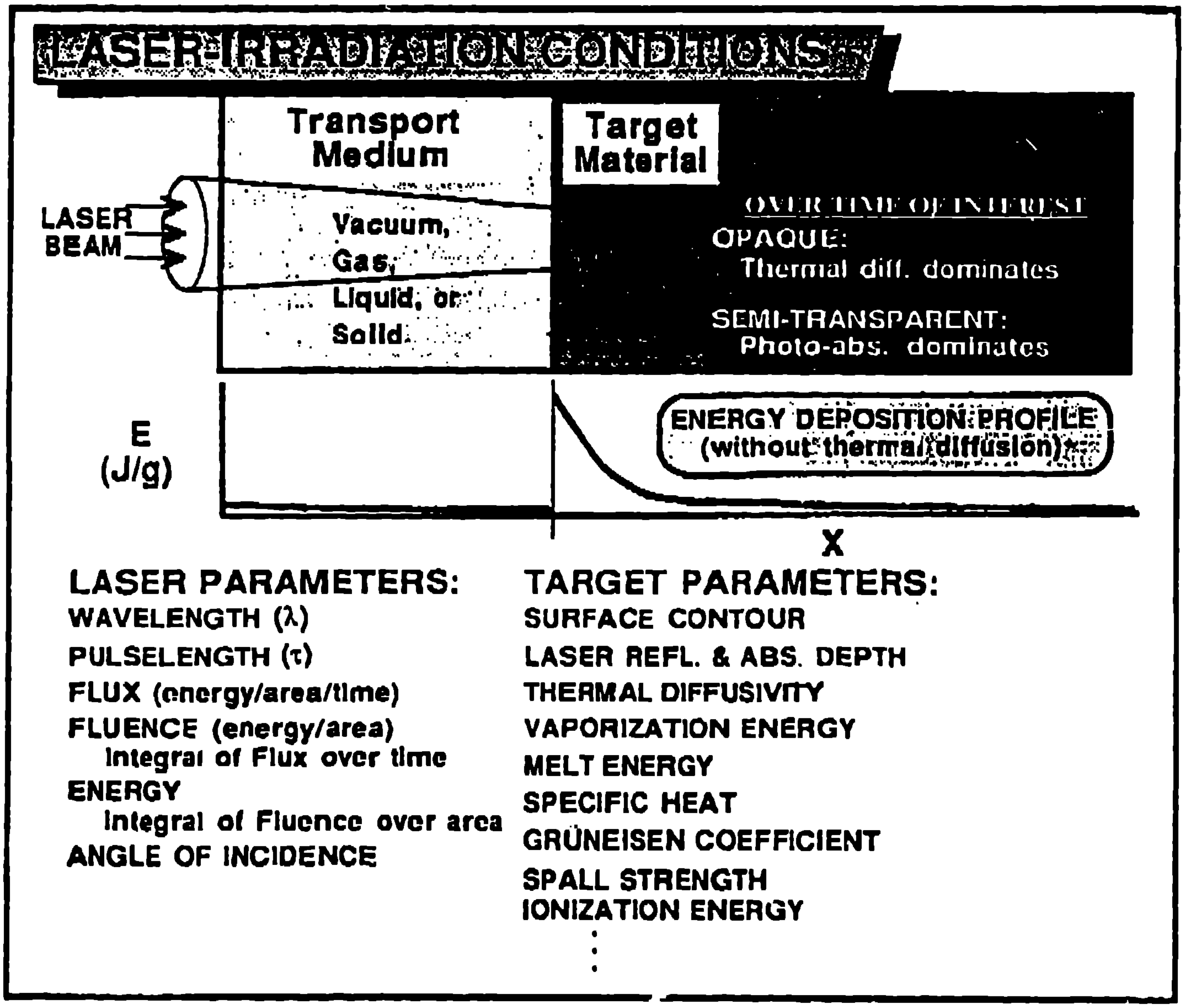

Figure 1. Laser irradiation conditions important to laser-ablation processes.

assumed that the laser beam irradiates the target material through some (essentially) transparent, transport medium. The target material can be considered to be either opaque, in which case the laser absorption depth is small in comparison to the thermal diffusion depth (during the time of interest, which is usually the laser pulselength), or, otherwise, semi-transparent. The ablation process is strongly dependent upon the transport medium in front of the target surface; if the medium is a liquid or solid, the expansion of the ablation products is greatly restrained, which has major implications.[1,2] At fluxes above some limit, which depends on the medium properties, the medium must be a vacuum to avoid ionization breakdown and blockage of the beam on the way to the target. The process depends on many parameters, including those listed in Fig. 1.[3] Near the threshold for ablation, the process is strongly dependent on the target parameters, however, the dependence weakens as the flux and fluence increase so that eventually the process depends essentially only on the ionization and plasma properties of the target material.

For the sake of discussion, in Fig. 1 the laser deposited energy density at the end of the pulse is shown

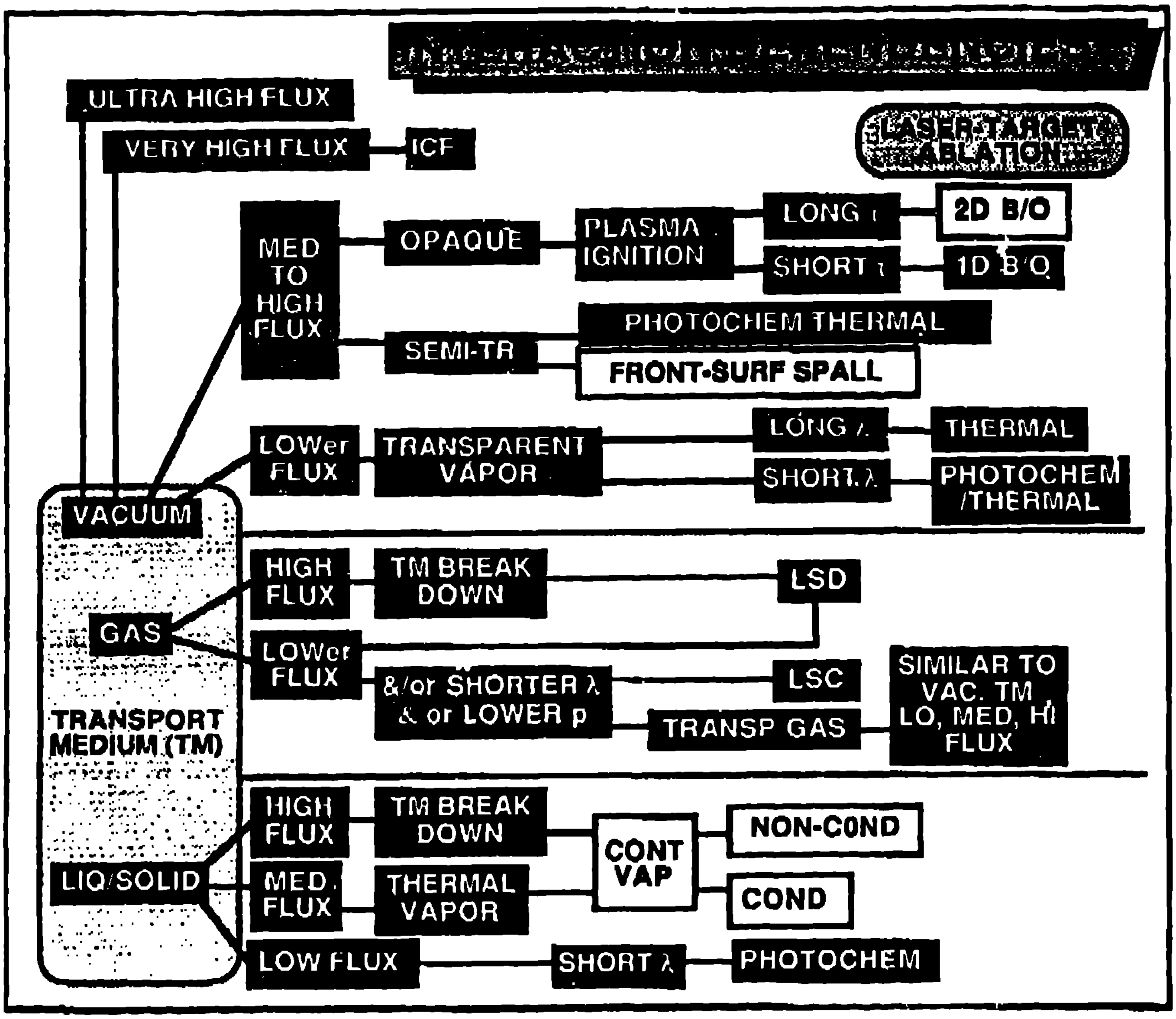

Figure 2. Interaction dependencies of laser-target ablation process.

as a function of position for a semi-transparent target (where thermal diffusion is negligibly small), for which the absorption coefficient remains constant during the pulse, producing an exponential deposition profile. Ablation can occur even when the front surface dose is below the threshold for vaporization, as discussed in Ref. 2. When the front surface dose is larger than the incipient vaporization enthalpy, ΔH_{iv}, vaporization will occur; non-vaporized material also is usually ejected along with the vapors--in part, because of phenomena associated with the fact that the complete vaporization enthalpy, ΔH_{cv}, is generally much larger than ΔH_{iv}.[1]

For opaque target materials, the laser absorption depth is negligibly small and the front surface temperature rises as the accommodation flux associated with thermal diffusion decreases with time until rapid vaporization begins, which cools the surface by taking away the latent heat of vaporization.[4] The rate of vaporization is principally determined by the net flux onto the vaporization surface.

3. INTERACTION DEPENDENCIES

The various categories of dependencies of the interaction of the laser, the target and the transport medium for ablation processes are illustrated in Fig. 2.[3] These dependencies will be discussed briefly, beginning with processes shown at the top of Fig. 2 and progressing downward.

There are several important dependencies on wavelength. For short optical wavelengths, the photon energy can be above the difference in quantum energy levels for molecular states, so that direct photochemicial transitions are possible. At high intensities multiphoton excitations can be important. At low intensities, excited states can sometimes decay by other means than photodecomposition, thus reducing the degree of ablation. For laser fluxes above the threshold for plasma ignition, the absorption cross section (generally, principally inverse bremsstrahlung) increases with the square of the wavelength and the free electron density (free electrons per unit volume) at the critical surface decreases with the square of the wavelength.

3.1. Vacuum transport medium

For irradiation of full density targets through a vacuum transport medium, at very-high (and higher) fluxes, large free electron densities occur near the front of the laser-target interaction region causing the laser photons to be absorbed at the critical surface where the plasma frequency equals the photon frequency, producing very large energy densities and consequent effects. This is generally the situation for inertial confinement fusion conditions because of the need to implode tiny capsules at high rates. At ultra-high fluxes, unusual multiphoton processes take place.

At intermediate fluxes, the blowoff will be ionized by the intense laser beam; after plasma ignition occurs, the blowoff will become opaque to the laser beam because of the large value of the inverse bremsstrahlung cross section and most of the photons will be absorbed before reaching the critical surface. This process is most prominent for opaque targets, where (even for targets with high thermal diffusivities) the energy density is sufficiently high to cause significant material vaporization and ionization. Analysis is particularly difficult for this process at intensities just above the threshold for ablation where (especially for long wavelength photons) the initial vaporized material has little ionization and is nearly (but not totally) transparent to the photons. As this blowoff absorbs laser photons, it is heated and begins to ionize, which increases its cross section for laser absorption so that it cascades to a level of ionization that is consistent with the irradiation flux; this process is referred to as plasma ignition. The time between the beginning of the laser pulse and plasma ignition decreases as the laser flux increases. After ignition, the blowoff generally becomes completely opaque to the laser beam and heats up to a temperature such that the thermal reradiation intensity approximately equals the laser intensity (thermal diffusion through the plasma is usually small compared to radiation transport in this region of parameter space). For relatively short laser pulselengths, where the blowoff traverses distances small compared to the laser beam diameter during the laser pulse, the blowoff can be assumed to move in a direction normal to the target surface; this greatly simplifies analysis such that simple modeling becomes reasonably successful at predicting behavior.[4]

However, for longer pulselengths, accurate, detailed analysis becomes much more complicated. For target exposures with axial symmetry, the process involves a two-dimensional (2D) expansion; otherwise the process is 3D. At intensities considerably above the ablation threshold, the blowoff material becomes ionized early in the laser pulse, and most of the rest of the laser energy is then absorbed by inverse bremsstrahlung over a sizable region in the blowoff, with few additional laser photons reaching the target. After blockage of the laser from the target begins, ablation of the target is sustained principally by reradiation from the hot plasma generated in the region where the laser is absorbed. There is a strong circular dependence between the various elements of the process, which makes accurate calculations difficult. For example, if the calculated hydrodynamic expansion is not accurate, then the location of laser absorption becomes wrong, which affects the radiation transport to the target, causing the mass ablation to be in error, which further degrades the hydrodynamics, etc. Because of this strong feed back between the different physical elements of the process, each of these elements needs to be done well to obtain accurate calculations. Experiments for these conditions using advanced diagnostics, including time-and-space-resolved laser-holographic interferometry and VUV and XUV spectroscopy, have been done using the Chroma Nd-glass, 1.05 µm wavelength, laser at KMS Fusion in Ann Arbor, Michigan with up to about 2000 J on target with pulselengths from 0.5 to 128 µs.[5] The objective of the laser-target interaction Chroma experiments was to establish confidence in modeling to reduce the uncertainty of impulse predictions in regions of parameter space where experiments can not be performed; the modeling details must be accurate to have confidence in predictions for these cases. Measurement of integral quantities, such as impulse, do not critically test the modeling. Thus to accomplish the overall objective, we deemed that it was worth extensive efforts to obtain data with comprehensive diagnostics, including time-and-space-resolved measurements of plasma behavior, to provide a sensitive test of the radiation-hydrodynamic modeling. There are many mistakes that can be made in modeling and still get the impulse about right; and. it is easy to fudge the codes to get the impulse right. But if the only test of the code is impulse, then there is little confidence in predictions outside the region of experiments. On the other hand, comparison with comprehensive diagnostics allows one to understand where the calculations are going wrong so that one can then either fix the problem or address how that error will affect predictions in specific application regions. After making significant modifications to the LASNEX 2D radiation-hydrodynamic code and its data base to account for the physics taking place in this regime, calculations are in reasonable agreement with these experiments.[6]

At similar fluxes for semi-transparent targets, a sizable absorption depth will cause the energy density to be less (than for opaque targets) but vaporization can occur either by thermal decomposition or, for short wavelengths, by photo-decomposition. Depending on the Grüneisen coefficient and the spall strength, if the pulselength is short enough, the ablation can occur by front-surface spallation,[2] in which case layers or fragments of material are ejected, in contrast to vaporization of molecules; this extends the ablation threshold to lower fluences, which means less heating of the target. Also, most of the deposited fluence is carried off in the fragments so that the temperature in the residual target material is much reduced compared to ablation by thermal vaporization. In this process, a (front surface) layer of material is heated before it can thermally expand causing a (positive) compressive stress pulse in that region whose amplitude can be calculated using the Grüneisen coefficient. This stress pulse will cause dynamic expansion, both toward the front surface and in the opposite direction. The stress pulse will be reflected at the front surface by the shock impedance mismatch producing a reflected (negative) tensile pulse, if the impedance of the material in front is less than that of the material. To have spall, it is generally necessary that the transport medium in front of the target material be a gas or vacuum and not a liquid or solid; otherwise, the shock impedance of the material in front will prevent a tensile wave of sufficient amplitude from developing. The tensile pulse trails the compressive pulse into the material, thus causing a characteristic bipolar stress pulse. If at some depth, the tensile stress exceeds the tensile spall strength of the material, then simple modelling predicts that the material will spall at that depth, creating a new boundary surface at which the stress becomes zero. The rest of the compressive pulse that is propagating toward the front surface will then be reflected from this new boundary. If the reflected tensile stress builds up to the spall stress again, then spall will occur again. Eventually, the criterion will no longer be met for another spall layer to develop and a residual tensile tail will be left in the material with an amplitude that is less than the spall strength of the material. If the back of the material is also a free surface, then the compressive pulse will reflect from there as a tensile pulse, which can cause back surface spall. Recent experiments[7] on water essentially confirm this predicted behavior except that, above a certain stress level, many cavitation sights at various depths were observed instead of spall planes; perhaps this has to do with the elastic-plastic properties of water being more complicated than assumed in the simple modeling discussed above.

At lower fluxes (and generally long pulselengths so that the fluence may be large), vaporized material will remain transparent, then ablation can occur by photo-decomposition at short wavelengths or thermal decomposition at any wavelength.

3.2. Gas transport medium

At high fluxes, break down will occur in the gas causing ionization, which will block the rest of the laser beam from reaching the target. After ionization occurs at a point in the gas, the deposited laser energy generates a shock wave that propagates away from the heated region. At high fluxes, sufficient ionization can occur at the shock front to make it opaque (by inverse bremsstrahlung absorption) to the laser beam causing a behavior similar to the detonation of high explosive where an energy source exists at a shock front, which prompts the name laser-supported-detonation (LSD) for this situation.[8] At somewhat lower fluxes, the beam will propagate through the (nominally transparent) gas without significant attenuation; however, ionization initiated at the target surface can cause ionization in the adjacent gas and generate a LSD wave that will propagate back toward the laser. Even for transparent targets, this ionization can be initiated at tiny imperfections or specks of dust on the surface; this process sets the practical upper limit for the allowable flux onto both refractive and reflective optics in a gas environment. Also, if the gas has aerosol particles suspended in it, these particles similarly can initiate the ionization so that the flux must be lower to propagate through such a gas. At shorter wavelengths and/or lower gas pressures and/or lower flux, the absorption cross section can be small enough to prevent a LSD wave from developing; then absorption occurs behind the shock front, more analogous to the manner in which energy is released during chemical combustion; accordingly, this process is referred to as laser-supported-combustion (LSC). When the mutually dependent values for the flux, wavelength and pressure are sufficiently small, then the gas will remain essentially transparent throughout the process; in this case the target interaction is similar to that already described for a vacuum transport medium with transparent vapor or a semi-transparent target. However, the vaporized material from the target will mix with the gas in front of the target as it blows off, which can significantly cool the blowoff as well as substantially increase the momentum because the mass of gas swept up by the blowoff can be large compared to the mass of the blowoff.

3.3. Liquid or solid transport medium

At high fluxes, break down will occur in a liquid or solid transport medium, which will block the rest of the laser beam from reaching the target. In this case, ionized vapors are generated in the break down region that are contained by the surrounding condensed material.

At medium fluxes, below break down, ablation of the target can occur by thermal vaporization at the interface between the target and the transport medium. The restraint of these vapors by the transport medium can enhance the amount of work done on the target by several orders of magnitude compared to the case for a vacuum transport medium.[1] The degree of this enhancement can be strongly dependent on the condensability of the vapors and the thermal conductivity of the condensed matter surrounding the vapor cavity. For condensable vapors, rapid energy transport to the cavity wall can occur by condensation, and if the wall has a large thermal conductivity, then this energy will rapidly diffuse into the wall, which can lead to essentially a vacuum in the cavity in a short time. For non-condensable vapors, work is done on the cavity wall by essentially an adiabatic expansion because the thermal conductivity of gas is small; in this case, the pressure in the cavity can remain sizable indefinitely.

At low fluxes, where thermal effects are negligible, ablation can still occur for short wavelength lasers by photochemical decomposition.

4. POTENTIAL APPLICATIONS

4.1. Front-surface spallation

Front surface spallation is a potential mechanism for laser ablation of materials that may have a number of significant applications, such as medical surgery.[2] Perhaps it could be used also for desorption or to launch material (for example, large biological molecules) from a surface into a region where other techniques might be used to study the launched material. For example, the launched material, whose stoichiometry should be well determined, might be vaporized in flight by exposure to another more intense laser beam for production of superconducting thin films by condensation on a nearby substrate. It might also have value in cutting or shaping materials, including precision etching of polymers, metals, ceramics or semiconductors.

4.2. Two-dimensional blowoff into a vacuum

A reasonable level of understanding exists for this process; however, detailed modeling involves complicated 2D radiation-hydrodynamic modeling. This modeling should be of value for helping to optimize the conditions used for certain potential industrial applications such as producing superconducting thin films. Because of the high temperatures, the ionization of irradiated material, and the short wavelength reradiation, all of which have potentially damaging effects on residual tissue; it would seem desirable to avoid this process, if possible, for most medical applications.

4.3. Contained Vaporization

Understanding the contained vaporization process seems important to laser-medical applications so that it may be avoided or minimized in some cases and perhaps utilized to advantage in others. For laser angioplasty, the aim is to remove undesirable tissue from the arterial wall and open the artery without further effects to the wall. With liquid in the artery during laser exposure, contained vaporization would significantly enhance the load to the arterial wall; however, this enhancement may not provide a significant aid in opening the artery. If possible, it might be desirable to introduce a gas into the artery at the point of laser exposure to eliminate the effects of the contained vaporization process. If gas is introduced, the damaging side effects from exposure might further be reduced by utilizing the front-surface vaporization process[2] to ablate undesirable tissue from the arterial wall. However, for laser lithotripsy, it may be desirable to maximize the work done by the contained vaporization process in order to provide the necessary energy to break urinary stones.

The contained vaporization process appears to be important in a number of other applications, some of which do not involve lasers; however, laser-simulation experiments could be of value is studying the process for these cases. Examples include reactor safety where steel cladding on UO_2 fuel rods might rupture, exposing liquid sodium coolant to UO_2 vapors; or an accidental beam dump at the Superconducting Super Collider where 400 MJ might be deposited in 300 μs in a less-than-1-mm-diameter column in the wall.[1] The details of the contained vaporization process are quite complicated. However, simple analytical modeling is useful for evaluating the overall effects. Detailed, quantitative experiments are possible but difficult. Quantitative modeling of the important non-equilibrium processes that can take place need further development.

5. SUMMARY

In summary, there are many different regions of parameter space in which vastly different laser-ablation processes take place. Because of the many parameters involved, and sometimes because of the inaccessibility of certain regions to experiment, modeling can be of value in guiding research and development for assessing the merits of various potential applications. A reasonable level of understanding of the basic physics involved exists in most regions of parameter space; however, adequate material properties knowledge is a frequent weakness in modeling. Confirmation of the modeling generally requires carefully planned control experiments with time and space resolved diagnostics along with material properties measurements. Of course, the final consideration for any laser application is the success of the empirical technique developed.

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