University of California, San Diego   **UCSD-ENG-100**

Survey of mechanisms for liquid droplet ejection from surfaces exposed to rapid pulsed heating

B. Christensen and M. S. Tillack

January 2003

Introduction

In recent years ablation due to rapid pulsed heating has been the subject of a considerable amount of research. Interest in the subject has escalated due to the growing number of scientific and manufacturing applications utilizing high-powered lasers. Researchers are generally interested in the mechanisms of ablation, the production and transport of vapor, liquid droplet formation and ejection, and material properties near the thermodynamic critical point.

While much of the recent research has concentrated on ablation as it is applied to manufacturing processes, the effects of ablation could also be significant in inertial fusion power plants. The presence of airborne particles, such as liquid droplets, could adversely impact the operation of an inertial fusion power plant. In this case target emissions would be the source(s) of rapid pulsed heating, and hence ablation. While the formation of vapor in IFE chambers may be removed, the presence of liquid droplets in an inertial fusion chamber could adversely interact with incoming laser or ion drivers, targets, or diagnostics. These droplets could also be radioactive, toxic and chemically reactive. Thus a characterization of the particulate in fusion systems is needed to better understand the safety hazard and operational limits that such material could impose. The effects of ion heating are not treated in any of the literature referenced herein. For inertial fusion power plants ion heating may be significant, and should be investigated further.

For manufacturing processes such as pulsed laser deposition of thin films (PLD), and the production of nanotubes and nanoparticles, a high power laser beam is utilized to evaporate and ionize the surface of a target. The products of the ablation are then utilized to form thin films, nanotubes, and nanoparticles. For these processes it is desirable that the ablation plume consist only of vaporized material; however, liquid droplets are manifest for many materials if the incident laser fluence is greater than a threshold value. Since liquid droplet ejection from surfaces exposed to rapid pulsed heating is observable in many of the manufacturing processes, and may represent a significant amount of process variability, it is vital to gain a sound scientific understanding of the phenomena.

Much of the relevant literature utilizes empirical methods to demonstrate the effect of laser fluence on the ablation rate. These empirical studies point to a distinct transition from one form of ablation to another at a threshold laser fluence [2-4]. The threshold laser fluence is a strong function of the material, and may be more pronounced for some materials than others. While there is a widely accepted theory that describes ablation below the threshold laser fluence, a unanimously accepted high laser fluence model does not exist. It is clear from empirical studies that the ablation mechanism dramatically changes at the threshold laser fluence. It is also apparent that liquid droplets are ejected from the surface while operating in the high laser fluence regime. The ejection of liquid droplets is responsible for the jump in the ablation rate that is observed; however, the mechanisms of droplet formation and ejection are in question. Note that while several theories exist concerning the mechanisms of liquid droplet ejection, no model exists for prediction of the ejected droplet rate, size or velocity.

Rapid pulsed heating is also of interest for determining the critical temperature of materials [3]. Rapid pulsed heating may allow further investigation into many fundamental scientific principles including, non-equilibrium phase change, superheating of liquids, and spontaneous nucleation of vapor in a superheated liquid, thermodynamic critical point, and hydrodynamic instabilities.

Mechanisms for liquid droplet formation and ejection

Several mechanisms may contribute to the ablation of a rapidly heated specimen. The energy flux of the source and the physical characteristics of the affected material may determine which mechanism occurs. Since mechanisms may also be temperature (time) dependent it is possible that ablation transitions between several mechanisms during an energy pulse. Some mechanisms could also occur simultaneously. A brief description of several possible mechanisms that are prevalent in the literature will follow. The models and empirical evidence that are given in the literature will be summarized in a later section. In this survey we have neglected fragmentation caused by the stress state of the liquid and concentrated only on thermal and pressure driven mechanisms of phase change.

Normal evaporation

Normal evaporation, or heterogeneous evaporation, is simply the escape of molecules or atoms from a liquid surface (region of high concentration) to the ambient gas in contact with the liquid surface (region of low concentration). Normal evaporation will occur whenever the vapor pressure in the ambient gas is less than the saturation pressure of the liquid at the liquid temperature. Since the saturation pressure of a liquid increases with increasing temperature, the rate of evaporation will also increase with temperature [1]. Evaporation from the liquid surface causes a decrease in liquid surface temperature.

Normal evaporation is easily described at temperatures below the boiling point of the liquid. The rate of atomic flux (atoms/m^{2}s) for this mechanism is given by [2]:

$$\dot{m} = \frac{p_s}{\left(2\pi m k_B T\right)^{1/2}} \tag{1}$$

where m is the mass of the evaporating molecule or atom, k_B is the Boltzman constant, and p_s is the saturation pressure at the liquid surface temperature T. The saturation pressure can be determined with the aid of the Clausius-Clapeyron equation [2]:

$$p_s = p_o \cdot \exp\left\{\frac{H_{lv}\left(T - T_b\right)}{RTT_b}\right\} \tag{2}$$

where p_o is the ambient pressure, H_{lv} is the enthalpy of vaporization, and T_b is the equilibrium boiling temperature at the ambient pressure.

Heterogeneous boiling

While evaporation and boiling each involve liquid-vapor phase change at a liquid-vapor interface, boiling includes the creation of vapor bubbles at discrete locations below the

liquid surface, whereas in evaporation the vapor escapes from the interface between the liquid and the ambient gas [5]. Once boiling occurs the temperature of the liquid remains approximately constant. This occurs because at the onset of boiling, energy that is added to the liquid is primarily used for the liquid-vapor phase change and not for raising the liquid temperature. Also the presence of moving vapor bubbles impedes the formation of a temperature gradient [5].

Heterogeneous boiling occurs when vapor bubbles are formed below the surface at a nucleation site. A nucleation site may consist of a scratch or pit at a solid-liquid interface in which gas or vapor is trapped, or at a foreign gas bubble existing in the liquid. When the nucleation site temperature exceeds the saturation temperature of the liquid, vapor bubble formation and growth may occur. It has been suggested that due to the short time scale involved and the probability of low nucleation site availability that heterogeneous boiling may not be important for short pulses [6].

Homogeneous boiling and Phase explosion

When a liquid is heated slowly the surface temperature-pressure relation follows Eq. 2. This heating process is shown in Fig. 1 as the binodal. However, if the heat rate is fast enough, the liquid may become superheated, that is the liquid temperature can exceed the boiling temperature. A superheated liquid is in a metastable state. If the temperature continues to increase, the spinodal (see Fig.1) is reached and the liquid becomes unstable and catastrophically relaxes to a liquid-vapor mixture.
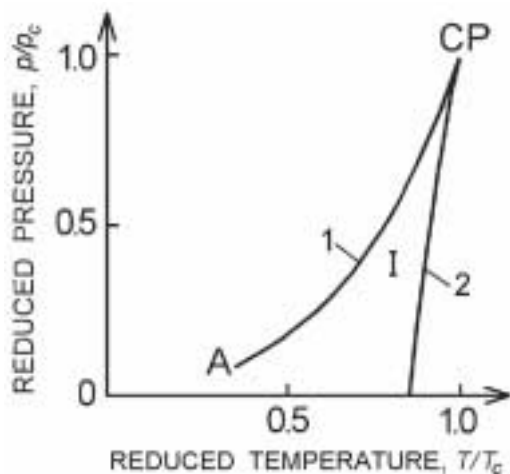

Fig. 1. A p-T diagram showing (1) the binodal, and (2) the spinodal. The area I is the metastable region. Given in [3].

Once in the metastable region a liquid need not reach the spinodal in order to change to a liquid-vapor mixture. A process that will prevent a liquid from reaching the spinodal is called homogeneous nucleation. It consists of the spontaneous creation of vapor nuclei within the liquid, without the aid of preexisting nucleation sites. Spontaneous nucleation

prevents significant superheating during slow heating processes. The rate of spontaneous nucleation is given by [2]:

$$J = \eta \exp\left(\frac{-W_{cr}}{k_B T}\right) \tag{3}$$

where W_{cr} is the energy required for vapor embryos to grow to nuclei. In order to reduce free energy, embryos smaller than the critical size will collapse, while those that are larger will grow and are considered nuclei. η is given by:

$$\eta = N\left(\frac{3\sigma}{\pi m}\right)^{1/2} \tag{4}$$

where N is the number density of the liquid and σ is the surface tension. Using this relation it can be shown that the rate of nucleation is small for temperatures less than $0.9Tc$ (where Tc is the critical temperature). It has been reported [2] that the frequency of spontaneous nucleation is approximately 0.1 s^{-1} cm^{-3} for temperatures near $0.89Tc$; the frequency increases dramatically to 10^{21} s^{-1} cm^{-3} for temperatures near $0.91Tc$. This suggests that for rapid heating processes a significant amount of superheating can be achieved since below $0.9Tc$ there will be no vapor bubble formation. Conversely, for slow processes there could be spontaneous formation of vapor bubbles well below $0.9Tc$, thus preventing superheating. The dramatic increase in nucleation frequency also portrays the catastrophic change of the metastable liquid to a liquid-vapor mixture.

Subsurface heating

Subsurface heating may occur when a thin layer of liquid is heated volumetrically, while the evaporation of atoms or molecules at the liquid-vapor interface removes heat from the liquid surface, thereby decreasing the surface temperature. The combination of evaporation and volumetric heating can result in the existence of a temperature maximum below the surface of the liquid.

This process could result in a catastrophic boiling crisis [3]. Since the rate of spontaneous nucleation dramatically increases near $0.9Tc$, even a slight temperature maximum could result in rapid nucleation and growth of bubbles below the surface of the liquid. As the vapor bubbles grow and coalesce, a large bubble may be formed below the surface. When the bubble reaches a critical size it will burst, propelling liquid droplets into the plume.

A similar result could occur for liquids that have a temperature maximum below the surface, but the maximum doesn't reach $0.9Tc$ *during* the energy pulse. In this case the rapid reduction of vapor pressure at the termination of the energy pulse may allow the liquid in the region of maximum temperature to become significantly superheated [3], resulting in spontaneous vapor bubble nucleation.

Hydrodynamic Effects

While the previous mechanisms dealt with phase change as the driving force behind mass removal, there are other possible mechanisms that are hydrodynamic in nature.

When a liquid is not heated uniformly, which may occur with a laser pulse, the variations in surface tension across the face of the liquid can create a hydrodynamic instability. If the acceleration of these waves is large enough, small liquid droplets could break off and result in the droplets being distributed in a radial fashion outside the heated zone [7]. Although this mechanism represents mass removal from the heated zone, it is expected that the velocity component normal to the surface wouldn't be significant. This makes this mechanism insignificant for engineering applications that are concerned with the ejection of liquid droplets normal to the surface, such as inertial fusion power plants, and PLD.

Another mechanism that is hydrodynamic in nature is stationary melt displacement and ejection. It has been suggested [8] that this process occurs during laser drilling of materials with high-powered lasers. The vapor pressure of the evaporating material is similar to a piston forcing molten material in the radial direction (see Fig. 2). This mechanism is probably not significant for ablation of *thin* films.
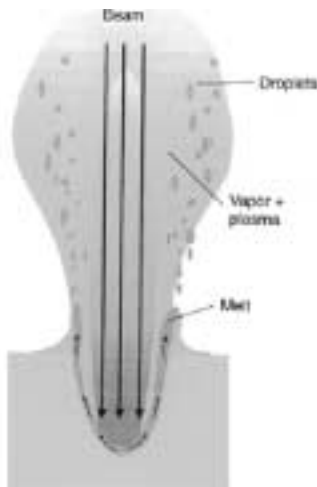

Fig. 2. Depiction of stationary melt ejection as given in [8]

Summary of important literature

While there have been a significant number papers published on this subject, only a few of the papers present unique information. Some of the references included are for historical value, while others are quite comprehensive in dealing with the mechanisms discussed above. It is suggested that the reader refer to the excellent treatment of this subject as given by Bulgakova and Bugakov [3]. Their paper is quite comprehensive, and contains many excellent references. A discussion of the findings from some important literature will follow. Because the primary objective of this report is to summarize the existing literature dealing with liquid droplet ejection, normal evaporation will not be discussed specifically. In addition the hydrodynamic effects will not be discussed

further, the interested reader should refer to [7] and [8] for a discussion of these effects and a list of other references.

Heterogeneous boiling

In many of the papers that deal with phase change mechanisms of liquid droplet ejection, heterogeneous boiling has been neglected. Miotello et. al. [6] suggest that the nucleation site density may be too low for significant heterogeneous boiling to occur. With only a small number of nucleation sites the nucleation and growth of vapor bubbles would not have the ability to produce the amount of liquid droplets observed during ablation.

Craciun et. al. [9] suggest that heterogeneous boiling may occur when a high fluence laser beam causes a "thick" liquid pool (i.e., the laser beam is not well absorbed by the material). The authors argue that neglecting heterogeneous boiling on the basis of a small nucleation site density [6] may be in error. This error is suggested to be a result of the constant nucleation site density proposed by previous authors. It is proposed that nucleation site density should be calculated as a function of superheat, which would increase the number of nucleation sites.

Three papers are referenced by Craciun et. al [9] which suggest that a bubble must reach a critical size before it can burst, and the liquid layer that confines the bubble must also reach a critical size. These three papers have not been reviewed for this work, but they might provide insight into droplet size for heterogeneous boiling or explosive boiling.

Craciun et. al. [9] question the explosive boiling theory on the basis that small liquid droplets ejected into a plume with superheated temperatures would have time to evaporate, therefore no droplets would be found on witness plates. Overall the proposals in Craciun et. al. [9] are interesting and offer an alternative view of what is taking place, but they offer no theoretical basis. Much of their thesis is based on comparing the surface morphology of materials irradiated by laser beams with wavelengths that are absorbed, to materials irradiated by laser beam that are poorly absorbed.

Homogeneous boiling and phase explosion

The majority of the recent literature dealing with liquid droplet ejection has focused on phase explosion. In the past it was generally assumed that subsurface heating was responsible for the high ablation rates during rapid pulsed heating as proposed by Dabby and Paek [10]. Miotello and Kelly [6] were among the first to suggest that phase explosion was a more likely cause.

Much of the evidence for phase explosion caused by rapid laser heating is based upon an observable jump in ablation rate when the laser fluence reaches a threshold value [2-4]. For these researchers the threshold fluence marks a transition from normal vaporization to phase explosion.

Bulgakova and Bulgakov [3] give an excellent theoretical and experimental treatment of this subject. They begin by giving the theoretical model for normal evaporation as a

function of the laser fluence. As given in Bulgakova and Bulgakov [3] the temperature distribution along the depth of a target is given by (in one dimensional form);

$$c_p \rho \left(\frac{\partial T}{\partial t} - u(t) \frac{\partial T}{\partial z} \right) = \frac{\partial}{\partial z} \lambda \frac{\partial T}{\partial z} + \left[1 - R(T_s) \right] \alpha_b I(t) \exp(-\alpha_b z) \qquad (5)$$

Where ρ is the mass density, c_p is the thermal capacity, λ is the thermal conductivity, and α_b is the absorption coefficient of the target. $R(T_s)$ the reflection coefficient which is assumed to be dependent on the surface temperature T_s. $u(t)$ is the surface recession velocity. If it is assumed that vaporized material flows according to the Hertz-Knudsen equation, and that the vapor pressure above the target surface is given by the Clausius-Clapeyron equation, then the surface recession velocity is given by:

$$u(t) = 0.82 \frac{p_b}{\rho} \left(\frac{m}{2\pi k T_s} \right)^{1/2} \exp \left[\frac{L}{k} \left(\frac{1}{T_b} - \frac{1}{T_s} \right) \right] \qquad (6)$$

Where L is the latent heat of the target, k is the Boltzman constant, and T_b is the boiling temperature at the reference pressure p_b. The intensity of the laser light reaching the surface of the target is given by:

$$I(t) = I_o(t) \exp \left[-\Lambda(t) \right] = I_o(t) \exp \left[-\int_0^\infty \alpha(n_p, T_p) dz \right] \qquad (7)$$

where $I_o(t)$ is the incident laser intensity, $\Lambda(t)$ is the optical thickness of the plasma plume, and α is the plasma absorption coefficient which is dependent on the plasma density and temperature.
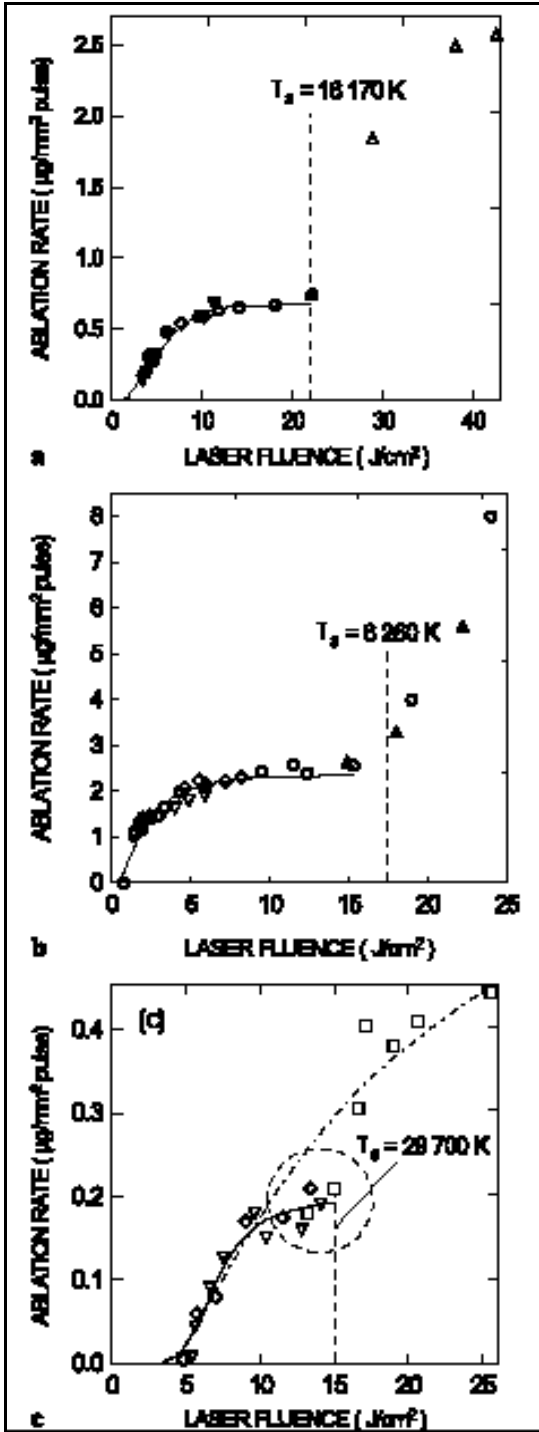
Fig. 3. Mass removal per pulse for (a) graphite (b) YbaCuO superconductor (b) and niobium (Taken from [3]). The solid line represents the model and the points are the experimental data.

Utilizing equations (6)-(8) along with initial conditions, boundary conditions and an approximation of $\Lambda(t)$, the mass removal rate is calculated as a function of laser fluence [5]. Experimental mass removal data are compared to calculated data for graphite, YBCO, and Nb in Fig. 3a-c. For each of the three cases the model and experimental data match well for laser fluences below the threshold value. At the critical laser fluence a sudden increase in mass removal occurs. Simultaneously a significant amount of liquid droplets appear in the ablation plume. These observations suggest a transition from normal vaporization to a more vigorous mass removal mechanism. The liquid droplets have been explicitly imaged as bright sparks [10], droplets have also been observed by placing a collection plate in the ablation plume [2]. The obvious change in ablation rate is attributed to explosive boiling. Notice that the transition from normal vaporization to explosive boiling is easily overlooked in Fig. 3c. This results from the low ablation rate for niobium.

Utilizing equations 6 and 7, the maximum surface temperatures are calculated as a function of temperature for each of the materials (see Fig. 4). Since the YBCO and graphite temperatures quickly approach a "saturation temperature" at high laser fluences, while Nb increases up to $0.9T_c$, it is assumed that Nb approaches T_c in a different manner. Utilizing this observation along with data for the time dependence of the surface temperature it is concluded that the difference in surface temperature behavior is not associated with plume absorption (see [3]). Utilizing the time dependence of the surface temperature and the plume intensity as a function of time, it is concluded that the screening of the surface, by the ablation plume, causes a saturation of the ablation rate.
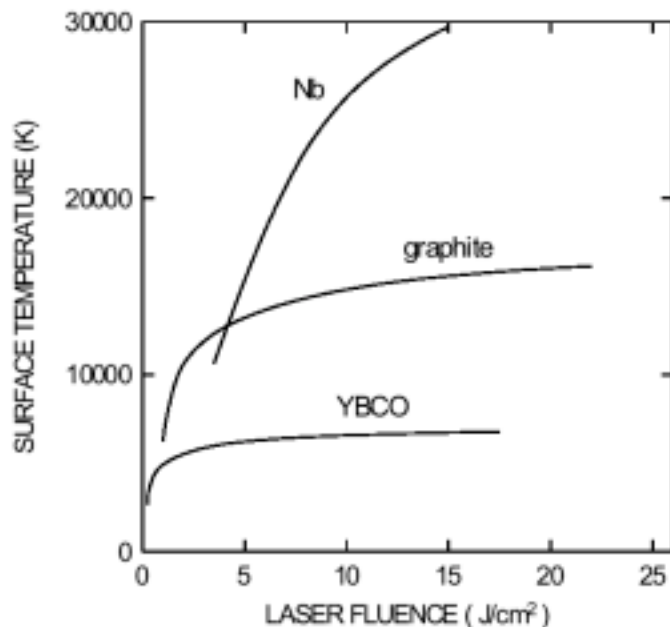

Fig. 4. The maximum surface temperature as calculated with equations 5-7. Taken from [3].

Xu and Willis [2] also reported similar results for laser ablation of nickel. While the same basic results are obtained, (i.e., normal evaporation occurs for laser fluences below the threshold value, while explosive occurs above the threshold value) some important points are included in this treatment.

Xu and Willis [2] point out that the properties of a material as it approaches the spinodal change drastically (see Fig. 5). These changes in material properties could affect the absorption of laser irradiation, the electrical conductivity, and the formation ejection of liquid droplets. The difficult task of obtaining properties such as surface tension and density near the spinodal may be required in order to accurately predict the ejected droplet size.
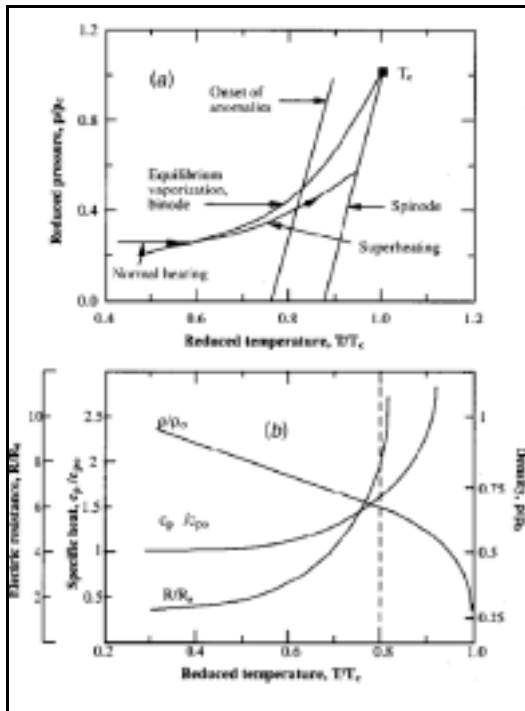

Fig. 5. (a) typical p-T diagram, (b) typical variation of physical properties. Given in [2].

In order for a nucleation to occur a vapor embryo must exist, and grow to a critical size. Once critical size is achieved the bubble will continue to grow in order to minimize free energy. However, a vapor embryo may also collapse in order to minimize free energy. The time lag for nucleation is the time necessary for an embryo to grow to the critical size. Equation 3, modified to account for the time lag is given as [2]:

$$J = \eta \exp\left(\frac{-W_{cr}}{k_B T}\right) \exp\left(\frac{-\tau}{t}\right) \qquad (8)$$

where t is the time for which the liquid has been superheated. The time lag is estimated as [2]:

$$\tau = \left(\frac{2\pi M}{RT}\right)^{1/2} \frac{4\pi\sigma p_s}{(p_s - p_t)^2} \tag{9}$$

where M is the molecular weight of the material. It is reported [2] that the time lag for metals is on the order of 1-10 ns. An apparent time lag is discovered by examining data for the vapor front velocity and the vapor plume transmissivity as a function of time.

Xu and Willis [2] utilize experimental data that shows a jump in vapor front velocity, vapor transmissivity, and ablation depth at the threshold laser fluence. Saturation of these values occurs for laser fluence above the threshold. The jump in these values suggests a transition from normal vaporization to some other form of ablation, namely explosive boiling. The saturation of the vapor front velocity also indicates a saturation of the surface temperature of the target. One may wonder what happens to the additional energy resulting from increased laser fluence, if a significant increase in surface temperature does not occur. Saturation of the surface temperature is attributed to the decrease in material absorbtivity as the spinodal is approached. This decrease in absorbtivity results in laser energy penetrating deeper into the material. An additional reason is that once nucleation is initiated, energy added to the system is used for nucleation and growth of vapor bubbles.

Miotello and Kelly [6] report that liquid droplets with dimensions exceeding the estimated dimensions of the superheated liquid (the superheated liquid is assumed to have a depth ~ 1/μ where μ is the absorption coefficient of the liquid). The dramatic increase in pressure over the liquid during explosive boiling is assumed to be the cause of this phenomenon.

Subsurface heating

Dabby and Paek [10] were among the first to propose subsurface heating as the mechanism of liquid droplet ejection. Miotello and Kelly [6] pointed out that the assumption of a constant vaporization temperature at the liquid surface was incorrect. From this Miotello and Kelly [6] concluded that subsurface heating would not be significant. However, Bulgakova and Bulgakov [3] determined numerically that subsurface heating could be significant for non-metal materials. As discussed earlier, subsurface heating could lead to results similar to explosive boiling. While these suggestions were made, no experimental data were obtained to verify the existence or effect of subsurface heating.

Conclusions and Recommendations

From the literature discussed above it is apparent that as the energy flux reaches the threshold value, ablation transitions from normal vaporization to some more aggressive mechanism. This threshold energy flux depends on the material, and the energy source. For instance it is reasonable that the interaction of a laser beam with a material could be quite different than the interaction of an ion with the same material. The exact details of

the aggressive material removal mechanism, such as the liquid droplet size, rate of liquid droplet ejection, and the material dependences, are unknown; however, there seems to be a growing consensus that explosive phase change is the cause.

Many engineering applications such as pulsed laser deposition, and inertial fusion power plants, will benefit from an increased understanding of liquid droplet ejection from rapidly heated materials. Future research could provide:
1. Prediction of the liquid droplet size
2. Prediction of the rate of liquid droplet ejection
3. Methods for controlling the liquid particle ejection
4. Insight into the effects of ion heating on liquid droplet ejection

References

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[9] Craciun et. al., 2002, "Laser-induced explosive boiling during nanosecond laser ablation of silicon," Applied Surface Science, 186, pp. 288-293.

[10] Dabby, F.W., Paek, U., 1972, "High-intensity laser-induced vaporization and explosion of solid material," IEEE Journal of Quantum Electronics, Vol. QE-8, no. 2, pp. 106-111.

Appendix

Brief outlines are given of the important papers referenced above. This may be useful for readers that are searching for information on a specific topic. This may or may not be included in the final report.

High-intensity laser-induced vaporization and explosion of solid material

1. Dabby, F. W.; Paek, U-C. (1972)
2. Propose that subsurface heating is the cause of liquid drop ejection.
3. In this case they are interested in how to increase the speed of laser drilling, therefore liquid droplet ejection is viewed as beneficial.
4. Important points
   a. Uniform heating of a liquid layer coupled with vaporization at the surface could result in a subsurface maximum temperature.
   b. This subsurface maximum could cause vaporization, which would increase the pressure below the surface of the liquid and eventually lead to an explosion.
   c. Report that material removal continues after termination of the laser beam.
   d. They utilize a "vaporization temperature". Below which they claim no vaporization occurs. Such a temperature doesn't exist.

Critical assessment of thermal models for laser sputtering at high fluences

1. Miotello, A.; Kelly, R.; (1995)
2. Propose that the model of subsurface heating was inaccurately derived, and that explosive boiling is probably the cause of liquid droplet ejection.
3. Important points
   a. Normal vaporization will occur from the extreme outer surface of the liquid for any temperature exceeding 0K. There is not a vaporization temperature as referenced above.
   b. Suggest that heterogeneous boiling may not be significant.
   c. Propose that phase explosion is the process by which liquid droplets are ejected.
      i. Liquid layer is heated to $0.9T_c$.
      ii. The effected zone makes a rapid change from a liquid to a liquid-vapor mixture.
   d. Pointed out that the spinodal is the thermodynamic maximum to super heating.
   e. Suggest that a dramatic rise in pressure over the liquid would occur with explosive boiling. This could lead to droplets whose dimensions exceed the depth to which the material is directly heated by the laser (This depth is related to $1/\mu$). Droplets larger than $1/\mu$ were reported.
   f. Proposed that subsurface heating is never important.

<u>Physical and material aspects in using visible laser pulses of nanosecond duration for ablation</u>

1. Koerner, C., et. al. (1996)
2. Experimental study investigating the depth per pulse, resulting geometry, affected zone.
3. The basic ablation process is classified into four main areas.
4. Suggest that the material and power density determine whether the ablation process will be by evaporation or melt ejection
5. Spallation is also suggested. Which occurs around a drill hole where material pieces are splintered off.
6. Present some results of short-time photography that show for large pulse energies the majority of the material is molten
7. Suggest evaporation is the main mechanism for low temperature processes.
8. Propose melt ejection for high power density pulses. In this case molten material is mainly expelled due to ablation and plasma pressure.

<u>Transport phenomena and droplet formation during pulsed laser interaction with thin films</u>

1. Willis, D. A., Xu, X.; (2000)
2. Transport phenomena and mechanisms for droplet formation in thin films.
3. Laser induced fluid flow caused by surface tension gradients.
4. Used to explain the appearance of droplets that are distributed radially outside of the affected zone.
5. High speed photography used to determine when the droplets form.
6. Suggest that the droplets form after the termination of the laser pulse.
7. Create a numerical model of the laser matter interaction.
8. Suggest that the recoil pressure is not a significant factor in mass transport.
9. Conclude that the laser beam creates a instability. If the surface wave acceleration is great enough small droplets may detach from the top of the wave.

<u>Phase change phenomena during high power laser-materials interaction</u>

1. Xu. X, Song, K. (2000)
2. Propose that explosive phase change is responsible for liquid removal.
3. Note that if the surface pressure is high enough, it will 'flush' the liquid out of the melt pool.
4. Utilize experimental results to show a transition from normal evaporation to explosive phase change at critical laser fluence.

     a. Jump in plume velocity beyond a certain laser fluence.
          i. Velocity of the plume increases for increasing temperature for normal vaporization
          ii. Velocity is relatively constant for increasing temperature once the phase explosion regime is reached.
     b. Transmissivity of the plume decreases for laser fluences up to the critical fluence, and then it remains relatively constant.

Non-equilibrium phase change in metal induced by nanosecond pulsed laser irradiation

1. Xu, X., Willis, D. (2002)
2. Propose phase explosion as the mechanism for liquid droplet formation.
3. Discuss the physics of metastable liquids
   a. Onset of material property anomalies.
   b. Spontaneous nucleation rate.
   c. Account for a time lag in spontaneous nucleation. This time lag is the time it takes for a vapor embryo to grow to a critical nucleus. This time lag is predicted, and confirmed experimentally
   d. Report that phase explosion has been attributed to picosecond laser pulses, yet the laser pulse time is much shorter than the time lag required for a critical nucleus to grow.

Pulsed laser ablation of solids: transition from normal vaporization to phase explosion

1. Bulgakova, N. M., Bulgakov, A.V. (2001)
2. Very comprehensive paper that deals with normal vaporization, explosive phase change, and subsurface heating.
3. Utilize experimental results to show the transition from normal vaporization to explosive phase change.
4. Point out that there is a small amount of liquid droplets present below the threshold fluence.
5. Suggest that subsurface heating could be significant for non-metal materials. Emphasize that subsurface heating cannot be neglected for many materials.
6. Suggest that a "boiling crisis" could occur at a subsurface temperature maximum where homogeneous nucleation rate would be higher. As the vapor bubbles form they may coalesce into a large bubble. As the subsurface pressure increases the film will eventually explode.