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Do, Nhan Thanh, M.S.

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Laser Ablation of Transparent Liquid Films on Opaque Solid Surface

A Thesis
Presented to
The Faculty of the Department of Physics
San Jose State University

In Partial Fulfillment
of the Requirements for the Degree
Master of Science

By
Nhan Thanh Do
May, 1992

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Abstract

Laser Ablation of Transparent Liquid Films on Opaque Solid Surface

by Nhan Thanh Do

An experimental study of the superheating of liquids on amorphous silicon (a-Si) thin film caused by a short ultraviolet excimer pulsed laser is presented to establish a primary understanding of the mechanism of the laser ablation of thin liquid films on opaque surfaces.

The temperature dependence of the optical constants for a-Si has been studied and it is observed that the extinction coefficient of a-Si increases significantly with temperature. This renders the measurement of the transient temperature at the liquid-solid interface during the laser pulse possible.

The transient temperature of the superheated liquid at the interface are monitored by a cw diode laser. To obtain the interface temperature, the nanosecond time-resolved transmission signal is calibrated through the application of the one dimensional diffusion model and the transfer matrix theory. The results are in reasonable agreement with the known data for superheated liquids in the literature as well as theoretical calculations.

To my parents, my brothers and sisters

To Luu Thao Chi

Acknowledgement

This work would have never been possible without much support, directly and indirectly, from many people. It is my great pleasure to take this opportunity to express my grateful thanks to them.

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I would like to thank Professor Peter T. Leung, a brilliant theoretical physicist from the Portland State University currently acting as a visiting scientist at IBM Almaden Research Center. I am indebted to his teaching ranging from the fundamental laws to the most recently discoveries in optical and thermal Physics, as well as the philosophy of this beautiful branch of science.

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Finally, I wish to express my deep gratitude to my parents, brothers and sisters for their ever-lasting supports and love. I would like to especially thank Luu Thao Chi whose love, encouragement, and understanding have helped me to keep the dreams alive.

San Jose, California
Nhan Thanh Do

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Chapter 1

Introduction

With the increasing importance of removing submicron particulate contaminants from substrate surfaces in the electronics industry, the technique of laser cleaning has been actively developed by various groups in the last few years [1 – 5]. This new development in laser cleaning is replacing the conventional cleaning techniques including ultrasonic and megasonics, wiping and scrubbing, high-pressure jets, liquid or gaseous etching, and even bombardment by fine particles [6]. There are currently many approaches in removing submicron particles by pulsed laser irridiation. J.D. Kelly and coworkers [4] have demonstrated that a particle adhering on a relatively transparent substrate that absorbs intense laser light can accelerate from the substrate by the sudden expansion of the particle itself. Laser cleaning can also be performed with a thin liquid film assisting on the substrate. By choosing the laser wavelength which is strongly absorbed by the liquid, S.D. Allen and coworkers [3,6] have shown that particulates can be removed with the explosion of the liquid film.

Concurrently, the IBM group headed by A.C. Tam [5,7] has been able to demonstrate that the most efficient approach in laser cleaning is to use a short-pulsed laser that is strongly absorbed by the substrate surface, coupled with the controlled deposition of a thin layer of a suitable liquid (of thickness on the order of microns) onto the particle-contaminated surface before the pulsed laser irradiation. In this technique, the liquid-substrate interface is heated up by the laser irradiation which then produces the superheating and explosive evaporation of the liquid film initiating at this interface. This technique can enable one to remove particles down to 0.1 micron in size from the substrate surface using a modest laser fluence of about 0.1 J/cm^2.

However, in spite of the success of the method, the detail of the mechanism of this approach has yet to be understood. It is expected that a thin layer of the liquid ($\sim 0.1 \mu m$) at the interface (See Fig. 1.1) must have been strongly superheated during the very short duration ($\sim 10\, ns$) of the laser pulse, and explosion then takes place with which particles are ejected from the surface by the huge liquid pressure due to explosion. Since the damage of the substrate surface is very sensitive to the temperature and pressure of the superheated liquid, the monitoring and control of these parameters become very important for both the understanding of the process and the protection of the substrate surface from damage.

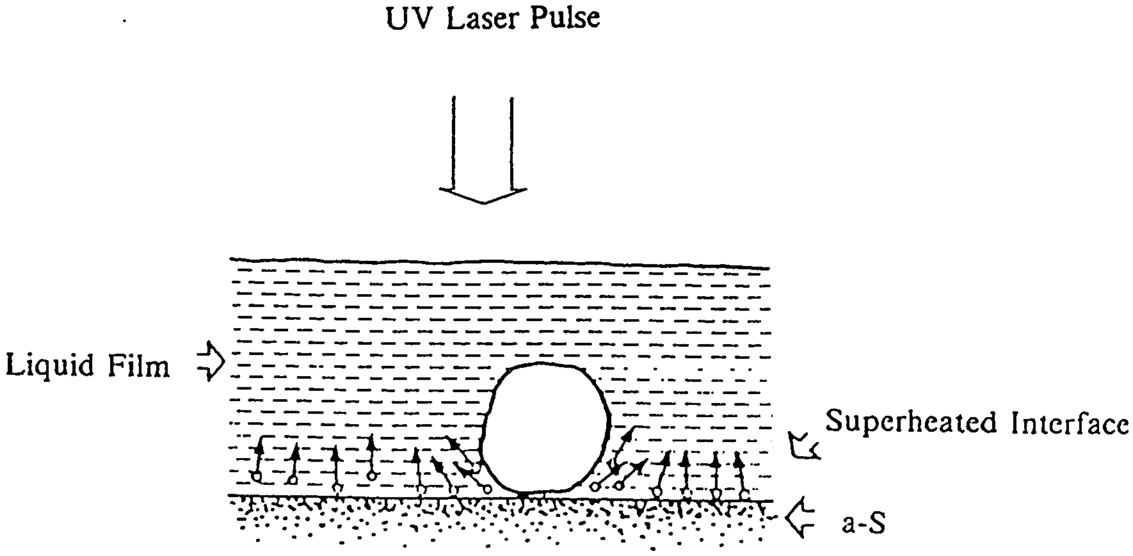

Figure 1.1: Illustration of laser cleaning with strong substrate absorption.

Besides the technological importance in the mechanism of laser cleaning, the thermal physics of superheated pure or mixed liquids has been an area of interest for many years. Questions such as the universal behavior, superheating limits, and possible supercritical phenomena concerning these metastable systems have been studied extensively in the literature [8]. It is well known that one of the efficient ways to create such metastable system is through pulsed-heating of the liquid. While pulsed electric current heating is usually achieved in the microsecond time scale, recent advances in short pulsed-laser technology can provide a much wider range of time scale for the study of the superheating of the liquids. In particular, sub-microsecond heating of the free surface of a liquid in a container as well as free liquid droplets [9] have been studied. In all these cases, the laser wavelength is in the absorptive region of the liquid and the heating is direct.

In this work, a study of the nucleation dynamics of the exploding liquid film is presented. The fast heating of the liquid is indirect. The KrF excimer laser with a wavelength of 248 nm and a pulse width of approximately 16 ns is used to irradiate an a-Si substrate surface with a transparent liquid film on top. The laser pulse is transparent to the liquid but strongly absorbed by the a-Si substrate which then heats up the liquid. This is called indirect heating of the liquid.

To monitor the change in temperature at the interface during the laser pulse, a cw diode laser at the wavelength of 752 nm is used to probe the change in transmission due to the heating of the solid-liquid interface. The transmittance drops when the temperature at the interface increases due to the change in the optical properties of the a-Si. A complete study of the temperature dependence of the optical constants for a-Si is included in chapter 2. Measurement of the transient temperature as well as the examination of the nucleation of the superheated liquid at the interface are presented in chapter 3. In chapter 4, the conclusion suggests some further possible investigations of the mechanism. In addition, experimental uncertainties and sources of errors are discussed in detail. Finally, references are listed to support the reading of this work.

Chapter 2

Temperature Dependence of Optical Constants For Amorphous Silicon

2.1. Introduction

The variation of the optical properties for solids with temperature has been studied extensively over the past few decades. Depending on the probe frequency and the substrate, this effect has been well studied for various metallic [1], semiconducting [2], and insulating [3] materials, with most of these being in their crystalline state. In particular, the optical constants n and k at elevated temperatures for crystal silicon (c-Si) have been measured in detail for different wavelengths [2]. The data from these measurements are obviously important in photo-thermal processing of the material, such as laser annealing. On the contrary, amorphous silicon (a-Si) is much less studied in this aspect, although there is an extensive literature on frequency dependence measurements [4]. Previous studies have reported that there is almost no temperature variation of the absorption coefficient for submicron thick a-Si samples at the Nd:YAG laser wavelength ($1.06\mu m$) [5 – 7]. It has then been sometimes assumed that unlike c-Si [2], the optical constants of a-Si can indeed be taken as independent of temperature [8].

In this work, we present a thorough study of the dependence of the real and imaginary refractive index n(T) and k(T) as functions of temperature T for a-Si. We shall see that such dependence is sensitive to sample thickness and optical wavelength, and can manifest itself significantly in high absorption regimes.

2.2. Experiment

The a-Si samples for our experiment are prepared by e-beam evaporation of c-Si in vacuum (10^{-7} Torr) onto fused quartz substrates ($250\mu m$ thick). The substrate temperature is kept at 140°C and the deposition rate at $10Å/s$. Different thicknesses of a-Si are prepared by controlling the deposition time; the uniformity and accuracy of these thicknesses are confirmed using the Tencor Alpha-Step 200 surface profilometer. The samples are then annealed at a temperature of 380°C in a nitrogen chamber for about 5 hours until optical measurements (reflectance and transmittance) versus change of temperature are reproducible for these samples. Under these conditions, the formation of oxide or nitride is kept at a minimum [9]. Stress relaxation of the sample is observed after the above heat treatment.

We have followed the standard reflectance ($\mathscr{R}$) and transmittance ($\mathscr{T}$) measurements for the determination n(T) and k(T) for our a-Si samples [10]. Two different wavelengths of light are incident at a small angle (5°) onto the sample which is mounted on a heated aluminum block (See Fig. 2.1). The heater-thermal couple system can vary the sample temperature from room temperature to about 400° C. The $\mathscr{R}$ and $\mathscr{T}$ signals at each temperature are detected by photodiodes which are connected to an oscilloscope. It is observed that for the 0.2 μm a-Si sample, both $\mathscr{R}$ and $\mathscr{T}$ are in the "interference region" [10,11] at the two wavelengths ($0.752\mu m$ and $1.15\mu m$) we studied. For the $1.0\mu m$ sample, clear interference patterns only show up for $1.15\mu m$, with $\mathscr{T}$ decaying monotonically with temperature for $0.752\mu m$ wavelength [11].

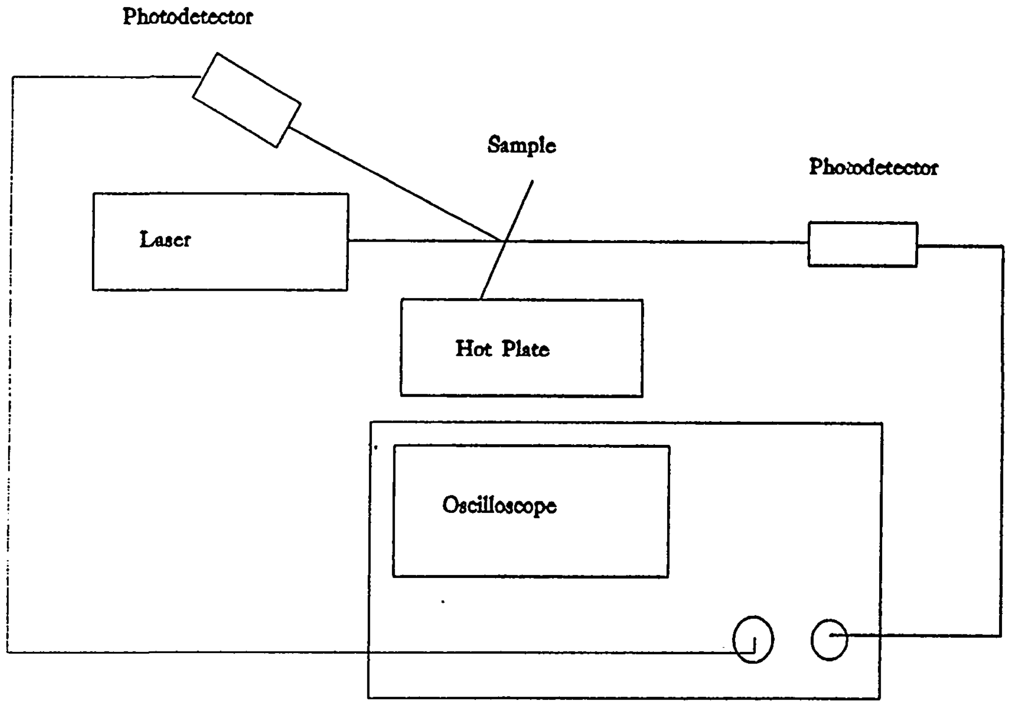

Figure 2.1: Experimental setup of the simultanuous reflection and transmission measurement.

2.3. Temperatrue Dependence of Refractive Index and Optical Gap Energy

The measured values for $\mathscr{R}$ and $\mathscr{T}$ are then fed into a computer iteration program to obtain values for n and k using Fresnel formula for a mutilayer system. The results are shown in Figures 2.2 and 2.3. In fitting the $\mathscr{R}$ and $\mathscr{T}$ values for n and k, we have taken into account the expansion of the layers by assuming the linear expansivity for a-Si and fused quartz to be $3 \times 10^{-6} {}^\circ C^{-1}$ and $5 \times 10^{-7} {}^\circ C^{-1}$, respectively. Figures 2.2a and 2.2b show the results for the $0.2 \mu m$ sample. While we see a slow linear rise of n(T) with dn/dT $= 1.3 \times 10^{-4} {}^\circ C^{-1}$, a situation similar to the case of c-Si, the behaviors for k(T) are quite different. At $\lambda = 1.15 \mu m$, we do get the average dk/dT ($< 10^{-5} {}^\circ C^{-1}$) totally insignificant, in agreement with previous observations at $1.06 \mu m$ $[5-7]$. However, k(T) shows a significant increase at $\lambda = 0.752 \mu m$ which is approximately linear with a slope equal to $2.3 \times 10^{-4} {}^\circ C^{-1}$. For the $1.0 \mu m$ sample, Fig. 2.3a shows the same linear behavior with dn/dT $= 5.0 \times 10^{-4} {}^\circ C^{-1}$ at $\lambda = 0.752 \mu m$. Fig. 2.3b shows significant increase of k with temperature, both at $0.752 \mu m$ and $1.15 \mu m$. While dk/dT $= 2.0 \times 10^{-4} {}^\circ C^{-1}$ for $\lambda = 1.15 \mu m$, k(T) for $0.752 \mu m$ is obviously nonlinear and can be fitted by the following expression :

$$k(T) = 5.48 \times 10^{-2} + 1.46 \times 10^{-4} T + 3.30 \times 10^{-7} T^2,$$

with temperature T expressed in ${}^\circ C$. The dependence of the optical constants on sample thickness for a-Si has been observed previously [5].

Thus we conclude that in the strong absorption regime, i.e., thick sample and/or absorptive wavelength, the absorption coefficient of a-Si can have a relatively strong temperature dependence. This could not be observed in the previous investigations simply because these previous studies were carried out in the highly transparent situation [5 – 7].

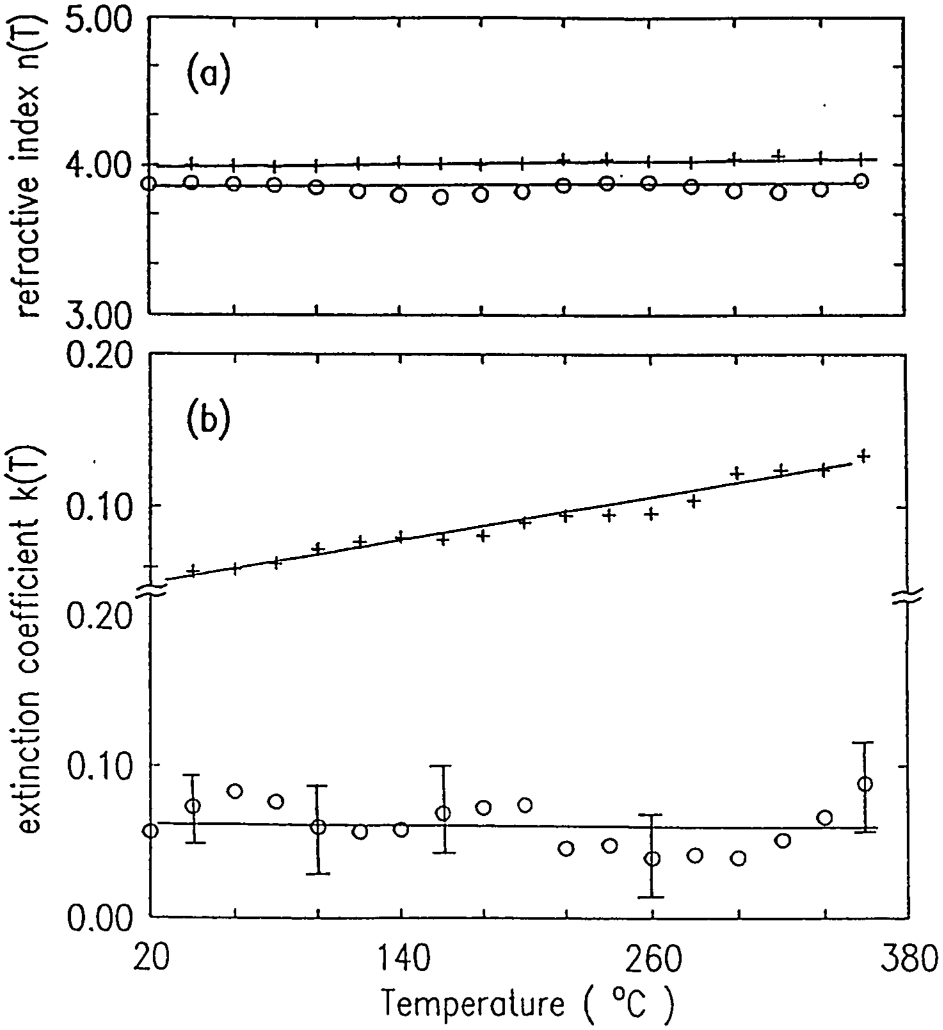

Figure 2.2: (a) Refractive index and (b) extinction coefficient as functions of temperature for 0.2 μm a-Si sample. (+)λ=0.752 μm; (O) λ =1.15 μm. The solid lines are from empirical fitting of the data. See text for details.

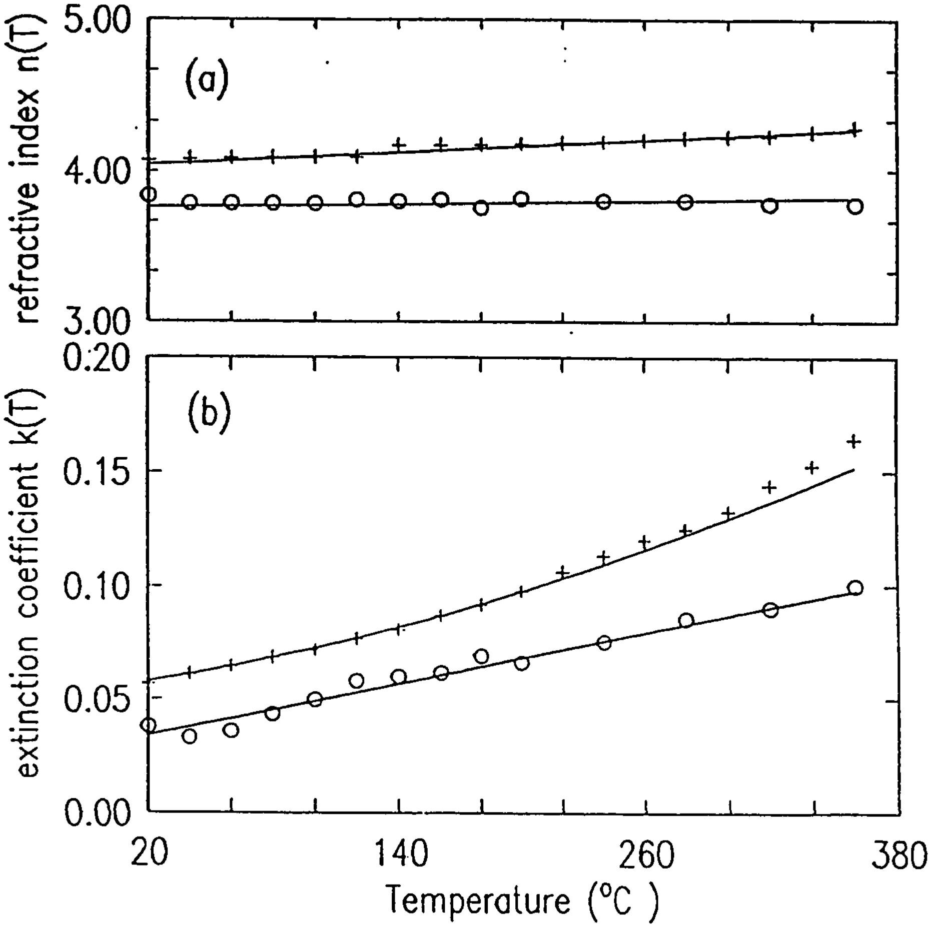

Figure 2.3: (a) Refractive index and (b) extinction coefficient as functions of temperature for 1.0 μm a-Si sample. (+)λ=0.752 μm; (O) λ =1.15 μm. The solid lines are from empirical fitting of the data. See text for details.

Furthermore, if we compare our results with those for c-Si, we observe that while dn/dT is comparable in both the a-Si and c-Si cases, k(T) generally increases much slower in the a-Si case compared to those in the c-Si case [2]. This may have to do with the very low mobility of the electrons in both the valence and conduction bands in a-Si as compared to those in c-Si [12].

In addition, it is of interest to note that by applying the Mott-Davis (MD) formula [12,13], it is also possible to deduce the variation of the optical gap energy (E_g) for a-Si with temperature. Although there have been other alternative forms put forward in the literature relating the quantities n, k, and E_g [14], the MD formula is still found to have high accuarcy for a-Si, both empirically [15] and theoretically [16]. Furthermore, it is not difficult to understand that as long as the temperature is much below the Fermi temperature of the substance, the MD formula should have good validity. In this approach, we have :

$$(n\alpha E)^{1/2} = C(E - E_g),$$

where $\alpha = 4\pi \frac{k}{\lambda}$ is the absorption coefficient, E is the photon energy, and C is a strength parameter. By plotting $(n\alpha E)^{1/2}$ vs E at room temperature for different wavelengths, the values for C and E_g could be determined at room temperature. We found that $C = 1.24 \times 10^4 eV^{-1/2} m^{-1/2}$ for the $0.2 \mu m$ sample and $C = 1.25 \times 10^4 eV^{-1/2} m^{-1/2}$ for the $1.0 \mu m$ sample, respectively. By assuming C stays roughly constant as the sample is being heated up, we can then use the results for n(T) and k(T) to determine E_g(T).

Figure 2.4 shows the result for $E_g(T)$ and the comparison with that for c-Si [17]. It turns out that all these curves can be fitted by the well known formula [18]:

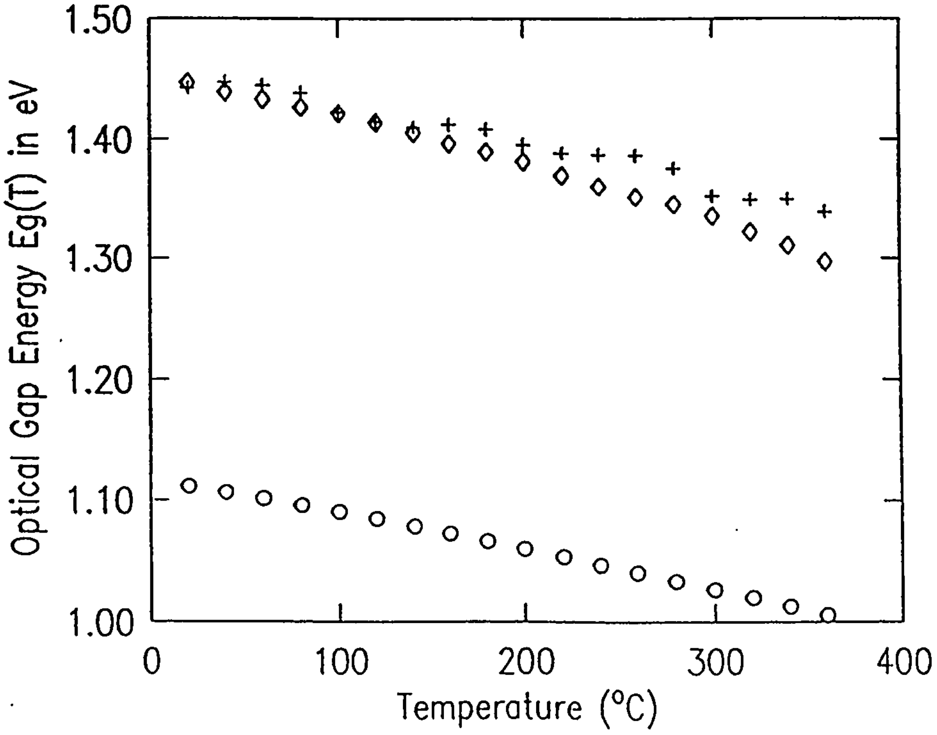

Figure 2.4: Optical gap energy E_g as a function of temperature for Si at $\lambda = 0.752\mu m$. (+) 0.2 μm a-Si sample; (◇) 1.0 μm a-Si sample; (O) c-Si (from Ref. 17).

$$E_g(T) = E_{g_o} - \frac{\alpha T^2}{\beta + T},$$

with E_g in eV, T in °K, α in eV °K^{-1}, and β in °K. The values for E_{g_o}, α, and β for both a-Si and c-Si are given in Table I. It is interesting to note that this formula for E_g(T) is widely applicable and can also fit the data for composite semiconductors [17].

Table I. Parameters for fitting E_g for both a-Si and c-Si.

Sample	E_{go} (eV)	α (meV °K^{-1})	β (°K)
c-Si[a]	1.155	0.473	635.0
a-Si (1.0μm)	1.508	0.665	669.7
a-Si (0.2μm)	1.495	0.494	646.6

[a] Values from Ref. 17.

We should also remark that only the results for 0.752μm wavelength have been used to determine $E_g(T)$ for a-Si, since for the 1.15μm case, the photon energy falls within the "absorption tail" in which the MD formula is no longer valid [12,15].

In summary, we have been able to measure the temperature dependence of the optical constants as well as the optical energy gap for a-Si up to 360°C. While we are aware that results for a-Si are in general very sample dependent (i.e., depending on the deposition conditions, the thickness, etc.), we expect that the qualitative temperature behavior for such physical quantities should have some general validity. This should stimulate a deeper theoretical understanding of such behavior in contrast with that for c-Si.

Chapter 3

Nanosecond Time-Resolved Transmission Measurement

3.1. Introduction

The thermal physics of superheated pure or mixed liquids has been an area of interest for many years. Questions such as the universal behavior, superheat limits, and possible supercritical phenomena concerning these metastable systems have been studied extensively in the literaure [1]. It is well known that one of the efficient ways to create such a metastable system is through pulsed-heating of the liquid. While pulsed electric current heating can usually be achieved in the microsecond time scale, recent advance in short pulsed-laser technology has been able to provide a much wider range of time scale for the study of the superheating of the liquids. In particular, sub-microsecond heating of the free surface of a liquid in a container [2], as well as free liquid droplet [3], has been studied in the literature. In all these cases, the laser wavelength is in the absorptive region of the liquid and the heating is direct. In this chapter, we present a study on the indirect superheating of a transparent liquid at the interface of an absorbing substrate surface in the nanosecond time scale. The motivation of the present work also comes from recent application of the huge pressure of these superheated liquids in technological developments as discussed below.

With the increasing importance of removing submicron particles (contaminants) from substrate surfaces in the electronics industry, the technique of laser cleaning has been actively developed by various groups in the last few years

[4 – 10]. This approach includes methods to heat up the particle [4], the substrate surface [5,6], as well as the liquid film [5,7 – 10] in cases when such a film is deposited on the surface to facilitate the cleaning process. Recent studies by A.C. Tam and coworkers [8] have shown that one of the most efficient ways to achieve laser cleaning of surfaces is to deposit a transparent liquid film (e.g., alcohol-water mixture) and to heat up the liquid-substrate interface with a UV excimer pulsed laser [7]. This technique can allow one to remove particles down to 0.1 micron in size from the substrate surface using a modest fluence of about 0.1 J/cm^2 [5]. However, in spite of the success of the method, the details of the mechanism of this approach have yet to be understood. It is expected that a thin layer of the liquid ($\sim 0.1 \mu m$) at the interface must have been strongly superheated during the very short duration ($\sim$ 10 ns) of the laser pulse; explosion then takes place and particles are ejected from the surface by the huge liquid pressure due to explosion. Since the damage of the substrate surface is very sensitive to the temperature and pressure of the superheated liquid, the monitoring and control of these parameters become very important for both the understanding of the process and the protection of the substrate surface from damage.

In this chapter, we study the nucleation dynamics of the exploding liquid film mentioned in the above process. In particular, data from the measurement of the liquid-solid interface temperature will be presented and compared with those for superheated liquids in the literature, as well as those from theoretical calculations. We have developed an optical transmission technique for such temperature measurements and have chosen a-Si as our substrate surface. The organization of the chapter is as follows. We shall start to first elaborate the rationale of our approach to this problem and the experimental details in Section 3.2, with results and analysis presented in Sections 3.3 and 3.4, and conclusions in Section 3.5. We shall see that in addition to the interface temperature, one can also give an estimate for the explosion time of the superheated liquid layer to be $\lesssim 1\mu s$, in general agreement with the values reported from experiments on explosion of liquid droplets [11].

3.2. Experiment

While there are many existing techniques for surface temperature measurement, optical reflection or transmission technique remains one of the most attractive options for its non-contact nature and fast time response capability. This technique is based on the variation of the optical properties of the substrate surface at elevated temperatures. Depending on the probe frequency and the substrate, this effect has been well studied in the literature for various metallic [12], semiconducting [13], and insulating [14] substrate materials. Although the effect for the reflection from the single air-substrate interface is generally very small (e.g., $d\mathscr{R}/dT \sim 10^{-5}K^{-1}$ for c-Si [15]), it can be much larger if the sample is deposited in the form of a thin film on an insulator substrate due to interference effects. One of the best known examples for the latter case is SOS (c-Si on sapphire), which has been studied extensively in the literature [16]. Also in this thin film case, both reflection and transmission can be monitored for a large range of probe frequencies. As for the single interface reflectance approach, most previous studies were limited to situations where there is phase transition (such as when the substrate surface is melted) in which a large change of reflectance (or transmittance) occurs. Since our expected range of temperature to be measured is somewhere from room temperature to about 400°C for the liquid (recall that the critical temperature of water is 374°C), which is far below the melting point of substrates like Si [15], we have decided to adopt the thin film interference approach. Previous investigations using this approach have achieved steady state [17] and millisecond [18] time resolution temperature measurements, in which uniform heating of the sample can be assumed. Bloembergen and coworkers [19] have studied picosecond pulse heating of SOS and the *average* temperature of the very thin (0.1 μm) c-Si film was obtained by considering nonuniform temperature distribution inside the Si film. In the following, we will consider a similar temperature profile effect for our nanosecond pulse heating and our main interest is to obtain the *surface* temperature of the substrate. More importantly, we believe that this present work is the first time that the dynamics of a heated liquid film is studied on such thin film structures using optical probing techniques [20].

Furthermore, we have chosen amorphous silicon as our substrate surface to study for the following reasons. As noted above, while stuctures like c-Si on insulator have been studied extensively for surface heating phenomenon, it seems that, to our knowledge, a-Si on insulator has not been studied for transient surface heating phenomenon, except when there is surface melting which may lead to recrystallization and occurs at a much higher temperature regime [21]. So it would be of interest to study transient heating of a-Si at a much lower temperature range and to compare the results with the case for c-Si. We shall see that to achieve the same elevated surface temperature, the UV laser fluence required is much lower in the a-Si case compared to c-Si. In addition, the preparation of a-Si film on insulating substrate is relatively easier and we expect that while the heating details (e.g., the interface temperature for a given laser fluence) will depend on the properties of the substrate surface, the nucleation dynamics of the liquid will be quite general on any opaque surface of comparable surface finish. Hence the results we obtain in this later aspect should have some validity if other kinds of substrate surface such as c-Si are used in the experiment.

There have been some indications in the literature [22] that the optical constants of a-Si are very insensitive to temperature change. This will render a transmission monitoring approach impossible since we look for the change of optical properties when the a-Si surface is heated by the excimer pulse. However, in a more thorough study [23], we have concluded that the previous results [22] are incomplete and are true only for an a-Si sample film probed at a highly transparent wavelength. With an optimization of these two parameters (sample thickness and probe wavelength), one can indeed see large temperature effects in the optical properties of a-Si. We have thus confirmed that our approach to the problem of transient surface temperature measurement for a-Si is indeed possible and, in fact, we shall use our previous steady-heating experimental data [23] for calibration of the present results in the following.

Our experimental setup is shown in Fig. 3.1. The sample is mounted with the KrF excimer laser (248 nm, ~ 16 ns) pulse incident at a small angle (~5°) and the cw probe diode laser (752 nm) going through it at normal incidence. In the case when reflectance is also monitored, the probe laser is incident at a small angle (~10°). The temporal evolution of the transmission signal is obtained by a fast photodiode and a digitalizing oscilloscope (HP 54111D). The energy fluence is determined by measuring the pulse energy using an energy meter (Molectron JD 1000) with the area of the UV beam measured to be ~ 0.5 cm^2. The liquid layer is deposited onto the sample using a homemade puffer which is described elsewhere [7]. The time delay (typically 0.1 s) between the puffer and the firing of the UV pulse is controlled by the timing generator, which first turns on the gas used in the puffer and then triggers the firing immediately afterwards.

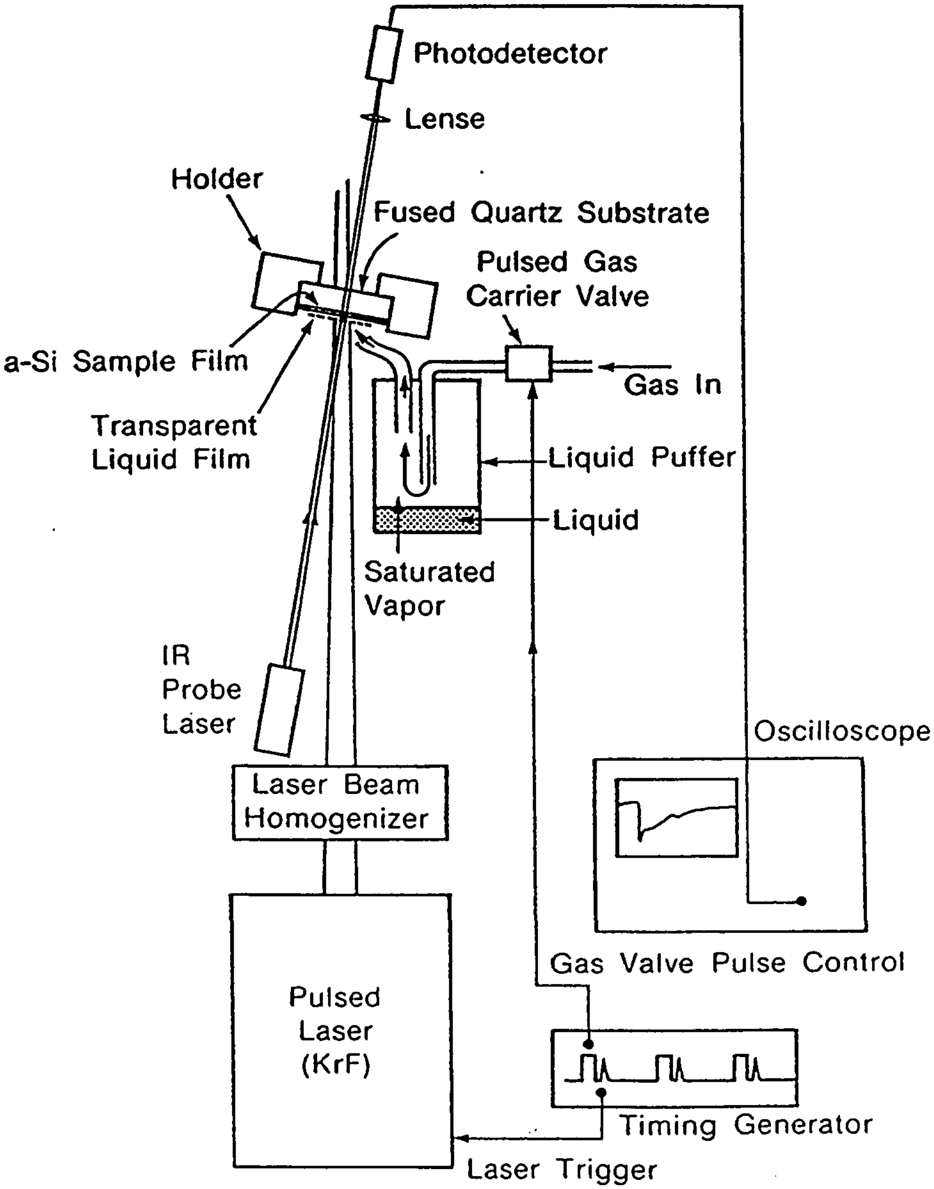

Figure 3.1: Experimental setup shown with the 752 nm probe laser and the 248 nm pulsed excimer laser.

3.3. Experimental Results

Some typical raw signals are shown in Figs.3.2 and 3.3 for two a-Si films of different thicknesses (0.2 μm and 1.0 μm) used in our experiment. Figure 3.2(a) shows the transmittance ($\mathscr{T}$) and reflectance ($\mathscr{R}$) of the probe diode laser which are complementary to each other, in response to the UV pulse irradiating at the bare (dry) surface of the 0.2 μm a-Si film. Figure 3.3(a) shows a similar behavior in $\mathscr{T}$ for the 1.0 μm film. Unlike the large transmission drop observed in previous experiments [16], which was due to the phase transition of the Si sample, these drops in $\mathscr{T}$ are due exclusively to the increase in absorption of the a-Si films caused by the rise in temperature. Figures 3.2(b) - 3.2(f) show the signals for a wetted surface with different liquids used. For simplicity, the $\mathscr{R}$ signal is only shown for the propanol case. In Fig. 3.2(b), the inset shows a long time evolution of $\mathscr{T}$ for the probe laser going through an isopropanol film (of microns thickness) deposited on the 0.2 μm a-Si film. Both the deposition and the evaporation phases for the liquid can easily be identified from the different periods in the inteference fringes. The arrow in the inset indicates the drop in $\mathscr{T}$ in response to the UV pulse. A similar one is resolved in the same figure in the nanosecond time scale together with the corresponding signal for $\mathscr{R}$.

From these signals, we can make the following observations: (1) the drop in $\mathscr{T}$ close to the end of the UV pulse indicates the temperature rise at the liquid-solid interface as in the case of bare surface; (2) an additional second dip in $\mathscr{T}$ appears for laser fluence above 17 mJ/cm^2; and (3) $\mathscr{T}$ and $\mathscr{R}$ are complementary to each other except at this second dip. Because the second dip appears in the cooling phase where the absorption ($\mathscr{A}$) in a-Si can only decrease [23], we must conclude that neither $\mathscr{R}$ nor $\mathscr{A}$ can account for this drop in $\mathscr{T}$. We thus interprete this second drop in $\mathscr{T}$ as due to diffuse scattering of the probe light at the liquid-solid interface. This occurs when the liquid undergoes spontaneous nucleation at a certain superheated temperature when the laser fluence gets above a certain threshold. This is analogous to the critical opalescence [24] observed in light scattering from liquids heated closed to the critical point.

Moreover, it is observed that the depth of the second dip in $\mathscr{T}$ increases with increasing laser fluences and it saturates only at relatively high fluence values, at which the a-Si surface may have been heated to a temperature much higher than that of the critical point of the liquid. We thus speculate two possibilities : (1) the pressure (P) or the temperature (T) or both these two parameters of the liquid may become supercritical [25] at the interface; or (2) both P and T remain sub-critical below a certain superheat limit [3,11,24] and the nucleation creates a very thin vapor layer which insulates the liquid from the a-Si surface. In addition, the time lag ($\tau \lesssim 1 \mu s$) for this second dip to occur is comparable to that observed in the droplet explosion experiments [11]. We attribute any difference to the fact that the present experiment involves heterogeneous nucleation as constrasted to previous homogeneous nucleation in droplet experiments [11].

We further observed that (τ) is generally independent of laser fluences. Table II shows the values of (τ) for different liquids on the 0.2 μm film, from which we see that τ for isopropanol is the longest among the liquids studied. Comparison of the values for different liquids leads us to conclude that for liquids of high surface tension such as water, the wetting of the a-Si surface is quite poor and the nucleation is highly heterogeneous [26]. This heterogeneous nucleation, is known to be more efficient than homogeneous nucleation which then accounts for the relatively short delay times (τ) for liquids like water. In fact, it can be seen from Table II that τ in general increases with the decrease of the surface tension of the liquids. In addition, we observe that τ is to a large extent independent of the UV laser fluence. Except for the case of pure isopropanol, explosion of the liquids is visibly observable for laser fluences somewhat above the spontaneous nucleation threshold. These are consistent with the established data in literature [27] concerning the pressure of the superheated liquids, from which one finds that except for isopropanol, all the other liquids we studied can be superheated to a temperature with a pressure well above one atmosphere.

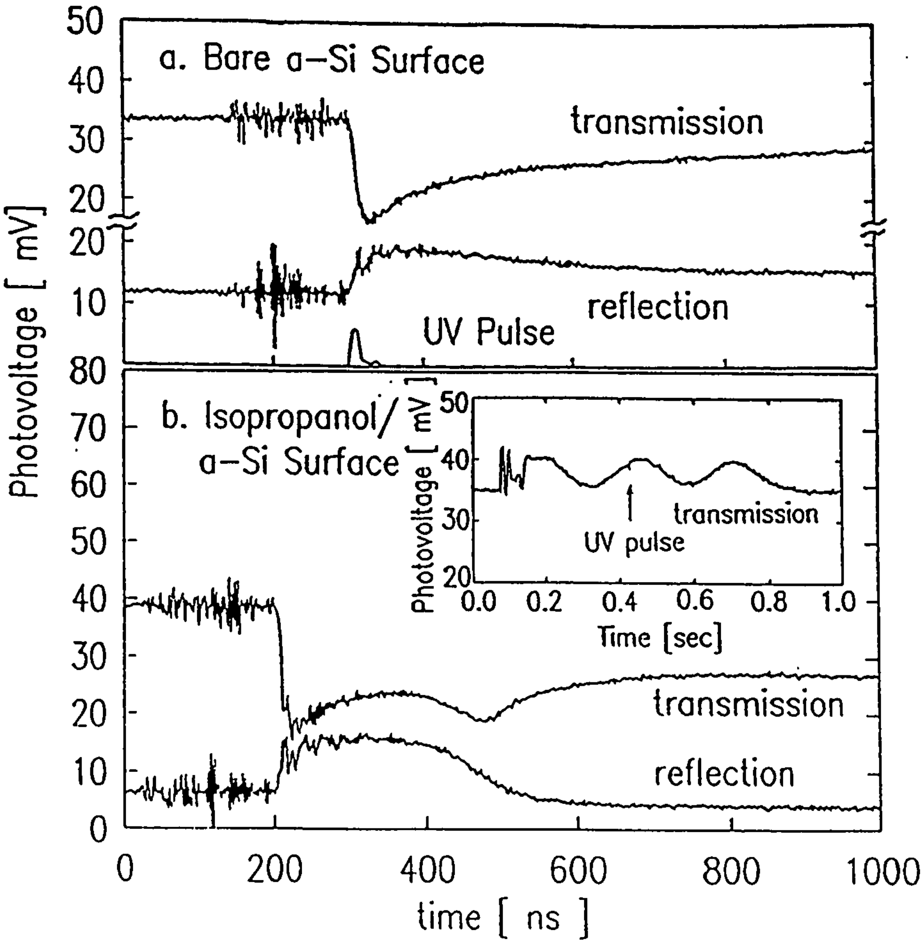

Figure 3.2: (a)-(b) Typical data for the probe transmission and reflection for the 0.2 μm a-Si sample.

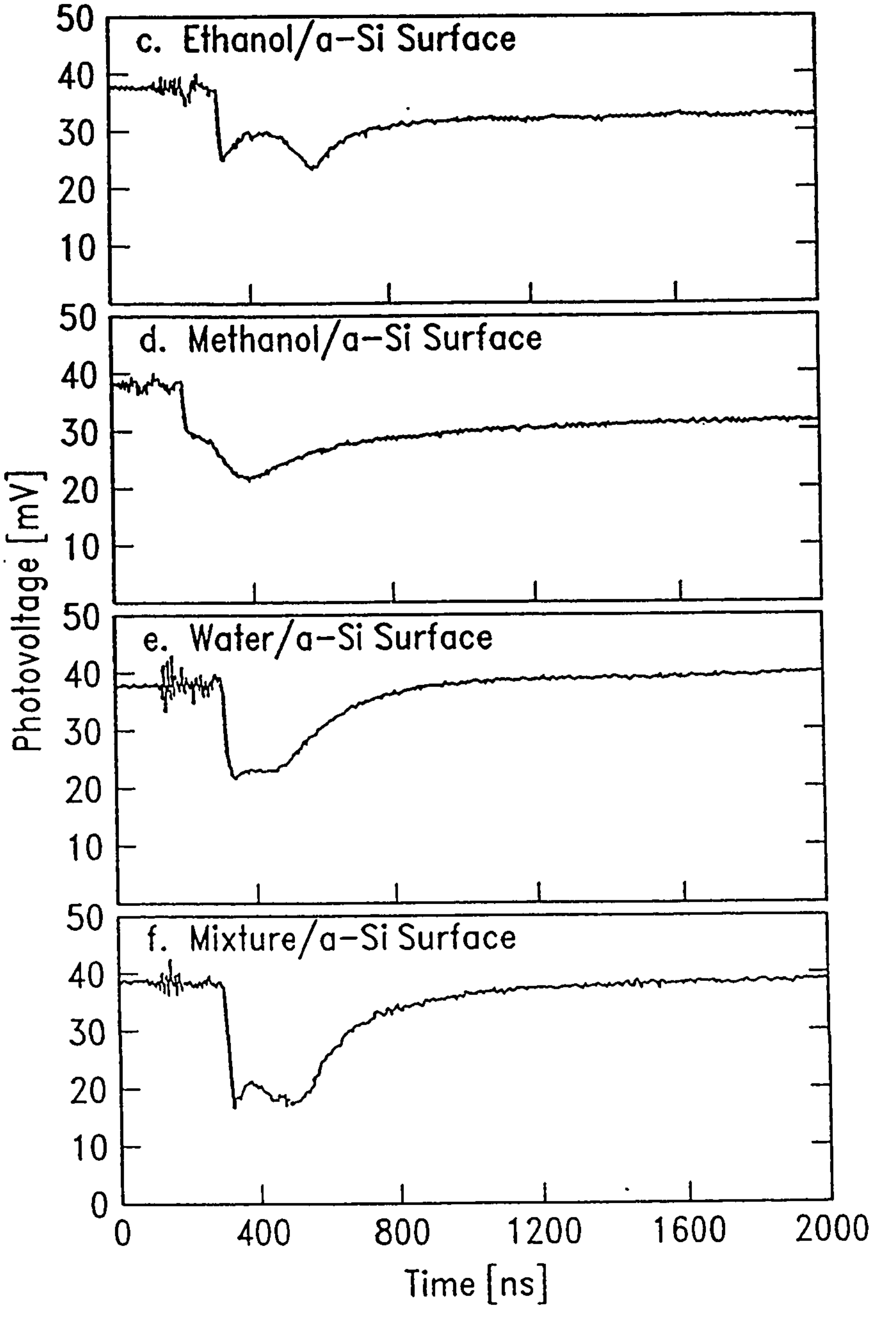

Figure 3.2: (c)-(f) Typical data for the probe transmission for the 0.2 μm a-Si sample. The "mixture" for Fig. 3.2(f) refers to 70% of water and 30% of isopropanol.

Table II. Time lag for spontaneous nucleation to take place after the UV pulse for different liquids.

Liquids	τ (*ns*)
Isopropanol	300
Ethanol	250
Methanol	250
Water/Isopropanol Mixture (70%/30%)	200
Water	150

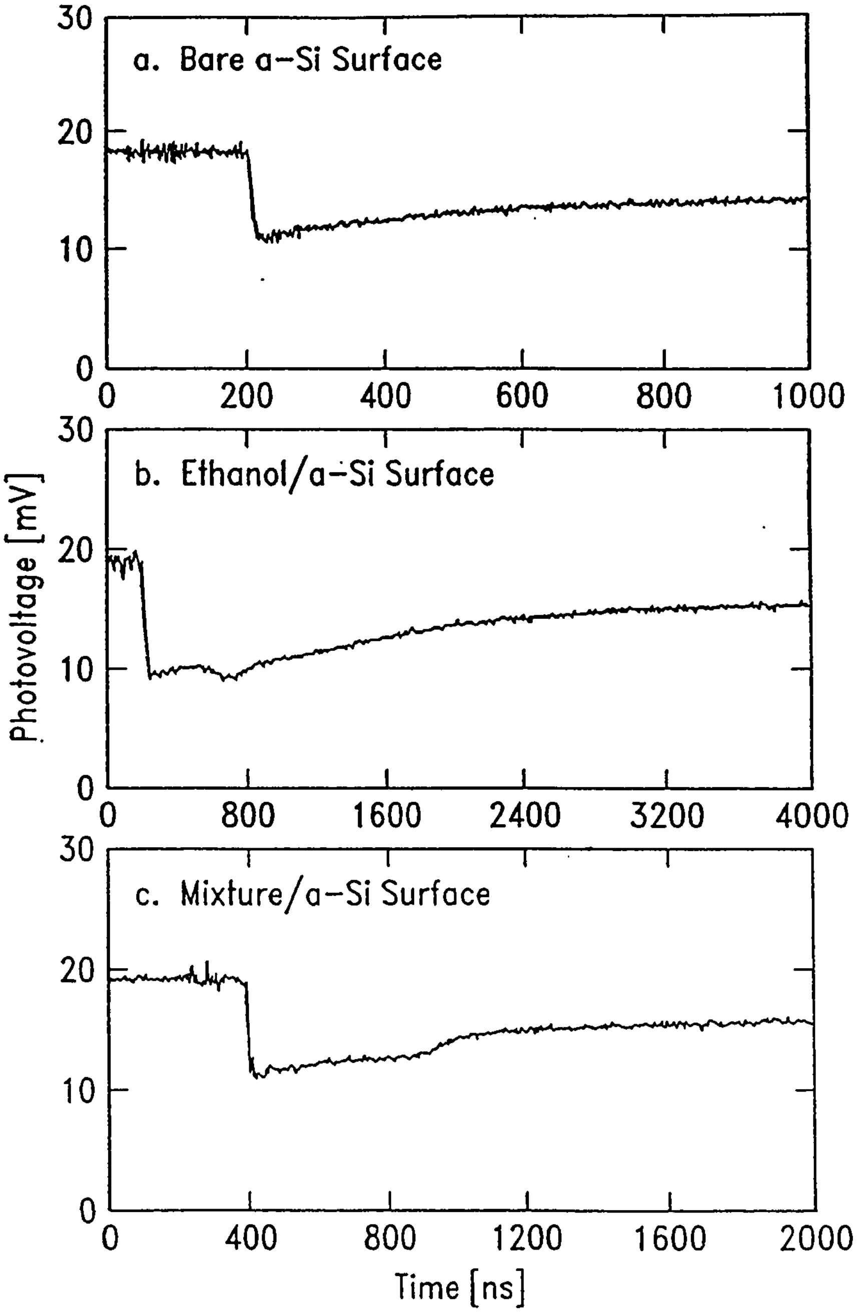

Figure 3.3: Typical data for the probe transmission for the 1.0 μm a-Si sample. Note the different time scales for each of 3.3(a), 3.3(b), and 3.3(c). The "mixture" in Fig. 3.3(c) refers to 70% of water and 30% of isopropanol.

Figures 3.3(a) - 3.3(c) show the corresponding raw data for the 1.0 μm sample. For the two liquids we have studied in this case, ethanol and isopropanol/water mixture, the nucleation signatures are in general less pronounced than those that appeared for the thinner (0.2 μm) sample. This is due to the fact that the thermal diffusion length is much shorter than the sample thickness in this case and the transmission signal actually measures an integrated thermal effect through the sample. For the same reason, the relative drop in $\mathcal{T}$ in this case is somewhat smaller for the same laser fluence. These points will be made more clear in the following when we come to the calibration of the results.

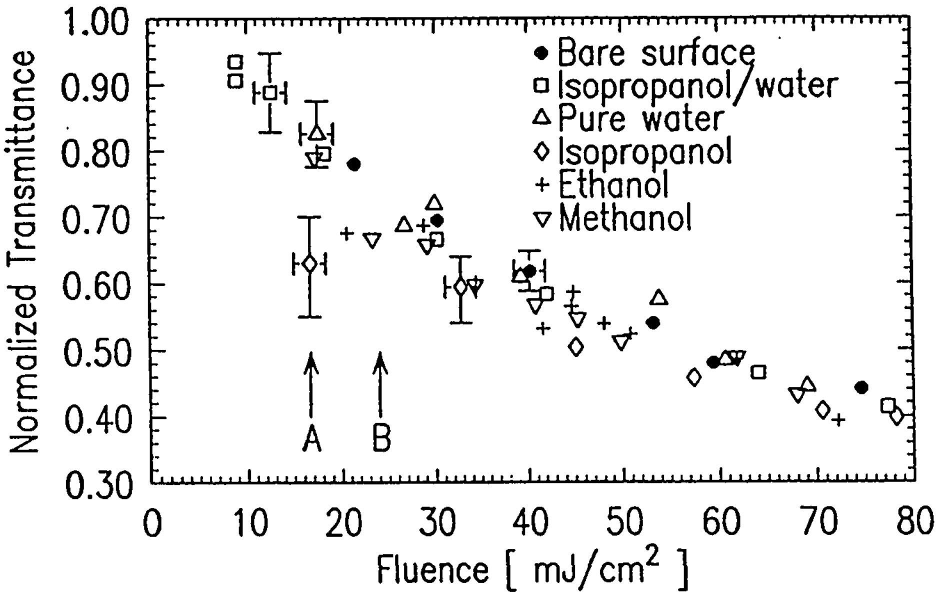

Figure 3.4(a): Normalized transmittance plotted against laser fluence for bare and wet a-Si surface of 0.2 μm film with different liquids used. The arrows A and B indicate the spontaneous nucleation threshold for isopropanol and the visibly detected explosion threshold for pure water film, respectively. In the pure water case, the threshold for nucleation occurs very close to the explosion threshold.

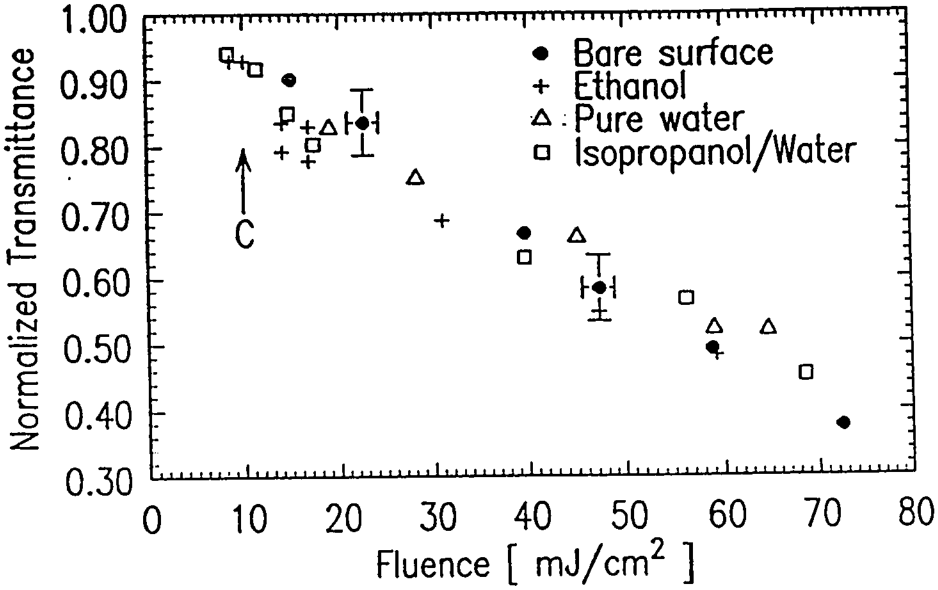

Figure 3.4(b): Normalized transmittance plotted against laser fluence for bare and wet a-Si surface of 1.0 μm film with different liquids used. The arrow C indicates the spontaneous nucleation threshold for ethanol film.

Figures 3.4(a) and 3.4(b) show a collection of all the data for the two samples of different thicknesses. We have plotted for various pulsed UV laser fluence the probe transmittance ($\mathcal{T}_n$) at the end of the UV pulse normalized to that at room temperature, i.e., before the UV pulse. The arrows A, B, and C in the figures indicate certain nucleation (second dip) and visibly observed explosive vaporization thresholds for some of the liquids as explained in the caption. For pure water, the two thresholds are practically indistinguishable. It is interesting to note that except for a few low fluence data for the alcohols, the results for $\mathcal{T}_n$ exhibit a universal behavior for the different liquids. Futhermore, we have performed experiments to study the effects of the liquid film thickness on our results. This is achieved by varying the delay time between the trigger for the puffer and the UV laser (see Fig. 3.1). This in turn controls the film thickness as can be seen from the inset of Fig. 3.2.

Figure 3.5 shows some results for $\mathcal{T}_n$ versus different delay times at a fixed fluence for isopropanol films on the 1 μm sample. As can be seen, with the delay time varied in a significant range, the measured $\mathcal{T}_n$ stays more or less constant within the error bars. Hence we can conclude that, within the present experimental accuracy, the liquid film thickness has insignificant effect in our measurement of $\mathcal{T}_n$. We believe that this reflects the fact that the reduced UV reflectance effect and thermal coupling effect due to the presence of the liquid film are roughly balanced out with each other, with the film quality not good enough to show coherent interference effect for the UV light. This can also be checked from a crude estimate using calorimetric arguments. Taking all these into account and also the fact that the UV laser is transparent to all the liquids we studied, we can conclude that to first order of accuracy, we have to consider only the a-Si film in the following photothermal analysis of our data for $\mathscr{T}_n$.

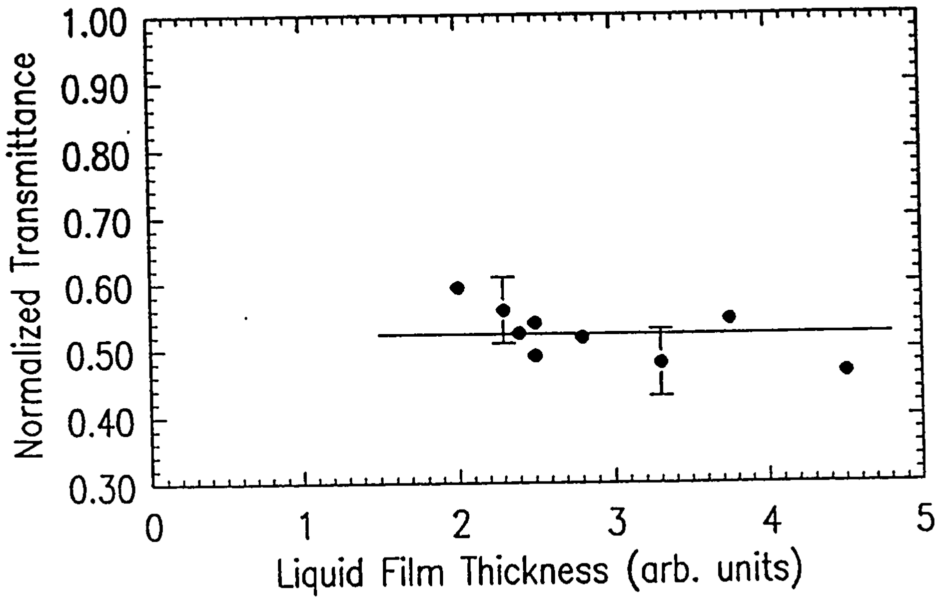

Figure 3.5: Normalized transmittance plotted against the different thickness (varied in a random fashion) of isopropanol film on a-Si surface at a fixed laser fluence. See text for details.

3.4. Calibration of the Transient Temperature at the Interface and the Nucleation Dynamics of the Superheated Liquids

In order to calibrate our results for $\mathcal{T}_n$ to obtain the interface temperatures, we have to take into account the effect of the temperature profile in the a-Si film due to the finite thermal diffusion length of the sample. Previously [23], in a steady-state experiment in which we have uniform heating for the a-Si film, we have obtained the temperature dependence of the optical constants (n and k) for the samples. To obtain the present calibration curves, we have to use these n(T) and k(T) to calculate $\mathcal{T}_n$ with the temperature profiles taken into account. To obtain these profiles, we resort to solutions from the one-dimensional heat diffusion equation:

$$\rho C_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial z}\left(K \frac{\partial T}{\partial z}\right) + Q(z,t). \qquad (1)$$

Here ρ, C_p, and K are respectively the density, specific heat, and thermal conductivity of the a-Si film, and $Q(z,t) = (1 - R)I_o(t)\alpha e^{-\alpha z}$ is the source function with R, α being the reflectivity and absorption coefficient for a-Si, respectively.

In order to solve (1), we have to know $I_o(t)$ which is the temporal profile of the UV pulse. Figure 3.6 shows a standard shape of our KrF laser pulse which we fit using the following equations for a triangular pulse shape :

$$I_o(t) = \begin{cases} \dfrac{2F}{t_1 t_2}\, t \,, & for \quad 0 < t < t_1 \\[2ex] \dfrac{2F}{t_2(t_2 - t_1)}(t_2 - t) \,, & for \quad t_1 < t < t_2 \end{cases} \,, \qquad (2)$$

with $t_1 = 6$ ns and $t_2 = 26$ ns, respectively. F is the fluence of the pulse and we have shifted the time origin to about 48 ns. The optical absorption coefficient for a-Si at the KrF wavelength (248 nm) is taken as $1.43 \times 10^6/cm$ [28], and the thermal constants are taken from the literature [29] from which we found very minor temperature dependence for these constants. We are thus justified to take the average values for these constants within the temperature range considered for solving Eq. (1) [30]. Substituting Eq.(2) in Eq. (1), we first calculate the temporal profile for T at the surface (z=0) and find that T peaks at about 15 ns after the incidence of the pulse. Then, assuming the deepest drop in $\mathscr{T}_n$ corresponds to the peak value of T(z=0), we then calculate T(z, t=15 ns) as our temperature profile for calibration purposes. After obtaining this profile, we then approximate it by many thin slabs of different temperatures T_i with the optical properties of each slab characterized by $n(T_i)$ and $k(T_i)$, as illustrated in Fig. 3.7.

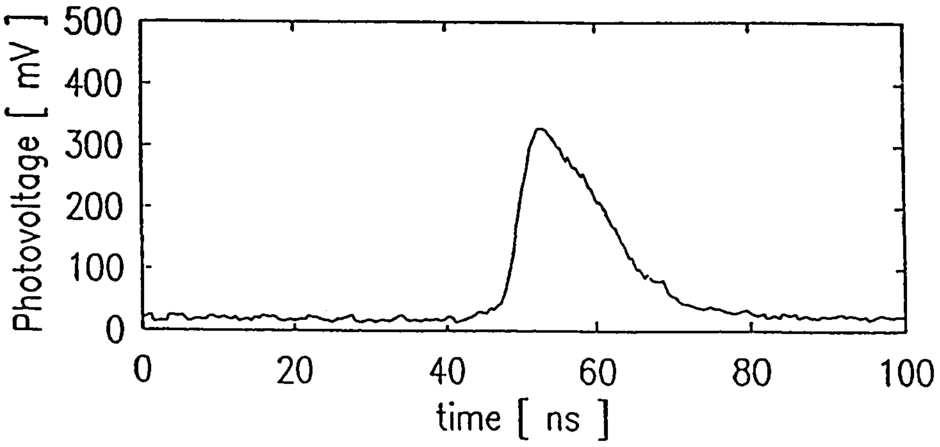

Figure 3.6: Temporal profile of the KrF laser pulse which is modeled as triangular in shape. See eq. (2) in text.

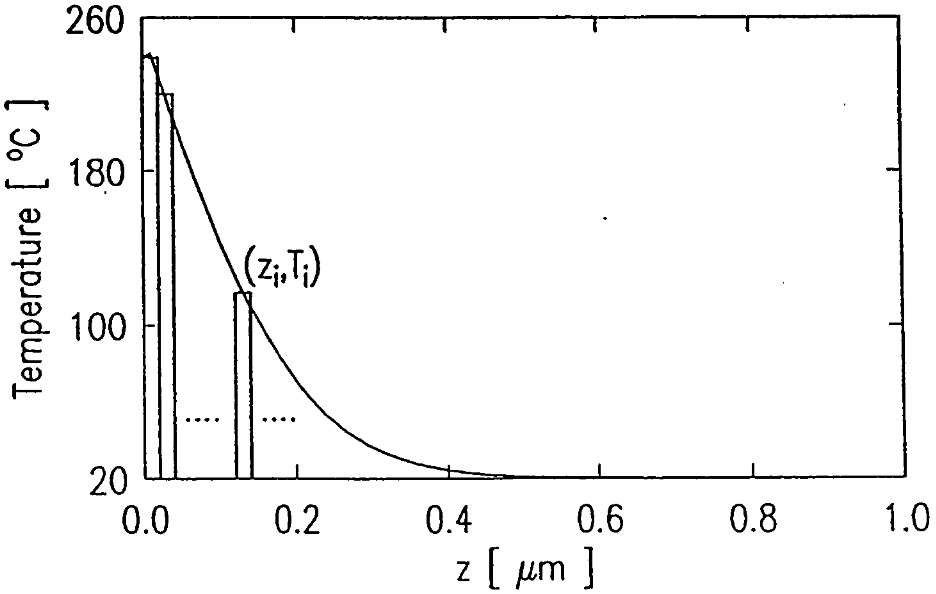

Figure 3.7: Temperature profile into the 1.0 μm a-Si film as obtained from solving the 1D heat diffusion equation.

To obtain the transmittance of the probe light through this collection of slabs, we start by writing down the transfer matrix for each slab as [31]:

$$M_i = \begin{bmatrix} \cos\beta_i & -j\tilde{n}_i \cos\theta_i \sin\beta_i \\ -\frac{j}{\tilde{n}_i}\cos\theta_i \sin\beta_i & \cos\beta_i \end{bmatrix}, \tag{3}$$

where $j = \sqrt{-1}$, $\tilde{n}_i = n_i + ik_i$, and θ_i is the angle of refraction in the *ith* slab; $\beta_i = \frac{2\pi\tilde{n}_i\Delta z_i}{\lambda}\cos\theta_i$, with Δz_i the slab thickness, λ the probe wavelength, and [23]

$$n_i = n(T_i) = \begin{cases} 4.00 + 1.30 \times 10^{-4} \times (T_i - 20)\ , & \textit{for the } 0.2\mu m \textit{ sample} \\ 4.07 + 5.00 \times 10^{-4} \times (T_i - 20)\ , & \textit{for the } 1.0\mu m \textit{ sample}, \end{cases}$$

$$\tag{4}$$

$$k_i = k(T_i) = \begin{cases} 4.60 \times 10^{-2} + 2.3 \times 10^{-4} \times (T_i - 20)\ , & \textit{for the } 0.2\mu m \textit{ sample} \\ 5.48 \times 10^{-2} + 1.46 \times 10^{-4} \times T_i + 3.30 \times 10^{-7} \times T_i^2\ , & \textit{for the } 1.0\mu m \textit{ sample}. \end{cases}$$

The overall transfer matrix is then given by

$$M = \prod_i M_i \equiv \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix}\ , \tag{5}$$

and the transmittance by :

$$\mathscr{T} = \left| \frac{2q}{(M_{11} + M_{12}q)q + (M_{21} + M_{22}q)} \right|^2\ , \tag{6}$$

with $q = \cos\theta_o$, θ_o being the angle of incidence. It is interesting to note that a similar profile analysis as the present one has been applied recently in studies of hydrogenated a-Si film where the profile for the absorption arises from the distribution of defects in the sample [32]. Figure 3.8(a) and 3.8(b) show our calibration curves obtained as described above for the two a-Si samples. We also show the result (the dotted line) in the uniform heating case for the $0.2\mu m$ sample obtained from the above fitted n_i and k_i. This result fits very well the nominal values of the data obtained in our previous steady state experiment, except that the interference pattern is washed away due to the fitting process.

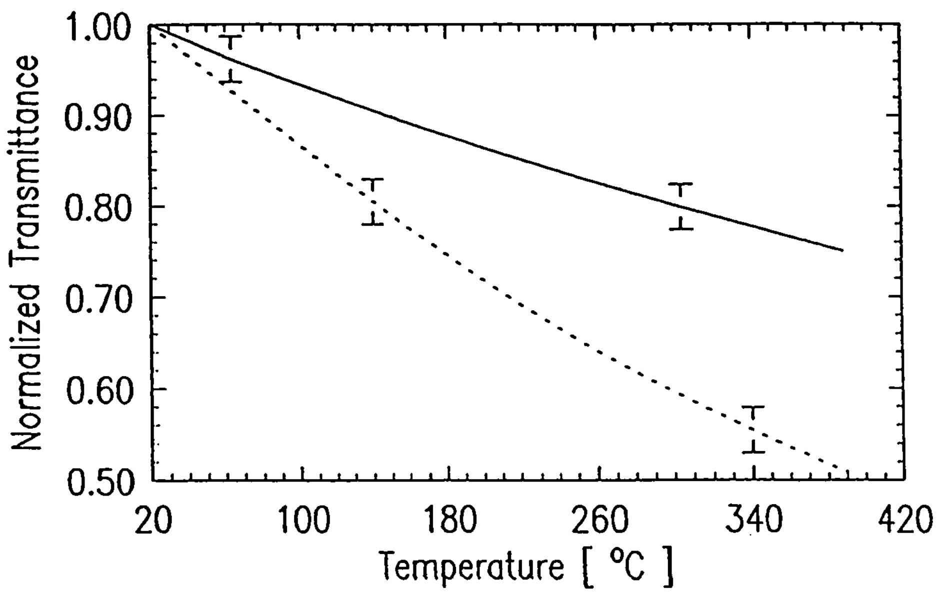

Figure 3.8(a): Normalized transmittance versus temperature curve obtained by applying the result fitted to our previous data from steady heating experiment (Ref. 23) for the 0.2 μm sample. The solid curve is the result converted for the transient calibration. The dotted curve shows the steady-state transmittance calculated from the fitted n(T) and k(T).

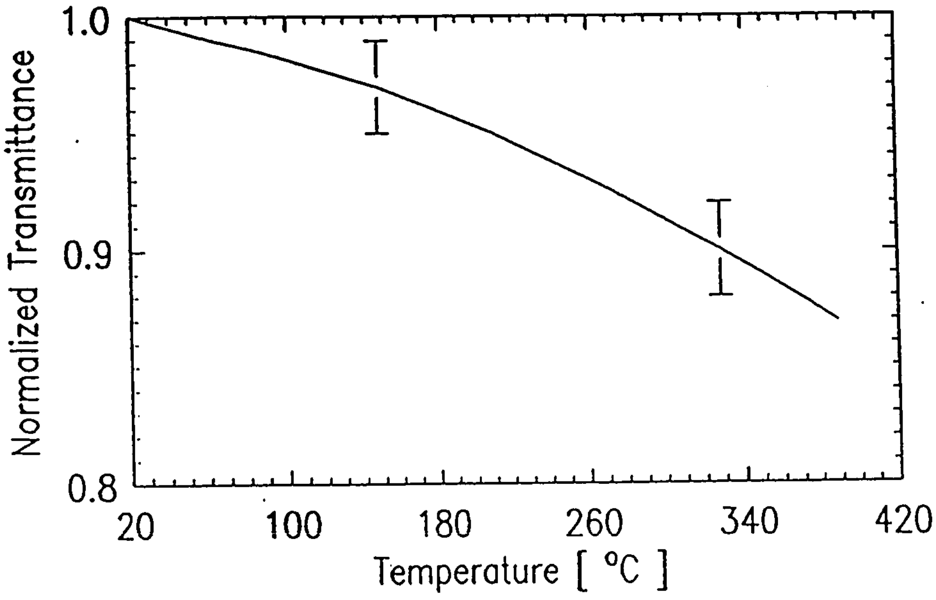

Figure 3.8(b): Normalized transmittance versus temperature curve obtained by applying the result fitted to our previous data from steady heating experiment (Ref. 23).

Using the solid curves in Figs. 3.8(a) and 3.8(b), the results in Figs. 3.4(a) and 3.4(b) are calibrated for surface temperatures below 400°C. This is so restricted since our steady-state experiment [23] was done only up to 400°C, for other complications such as oxidation and polycrystallization will start at higher temperatures [33]. Nevertheless, this range of temperature is enough for us to study the superheating of the liquids. Figures 3.9(a) and 3.9(b) show the calibrated results for the two samples with the solid lines showing the theoretical calculations for the peak surface temperature of an infinite thick a-Si sample. We observe that within experimental error, most of the calibrated temperatures are in reasonable agreement with the calculated results. Two data points for isopropanol show strange behavior at low fluences as can readily be seen from Figs. 3.4(a) and 3.4(b). We suspect that this may be due to the small signal to noise ratio at low fluences together with possible additional noise due to the fast evaporation rate for the heated alcohol. We also note that the agreement in the case for the 1.0 μm sample is not as good, and we account for this by referring to the larger temperature profiling effect in this case.

The many approximations (such as in solving Eq.(1) and in fitting n_i and k_i) introduced in the above profiling analysis simply leads to greater inaccuracy in the thick sample case. In addition, the threshold nucleation fluences from Figs. 3.4(a) and 3.4(b) are also indicated and they have values as 17 mJ/cm^2, 24 mJ/cm^2, and 10 mJ/cm^2, respectively, for isopropanol, water, and ethanol films. If we refer to the solid curves, we obtain the corresponding temperatures to be 210°C, 290°C, and 140°C, respectively. These are in reasonable agreement with the data from the literature which have values approximately to be 220°C, 307°C, and 195°C for the three pure liquids [27]. We should also remark that our identification of the deepest drop in $\mathscr{T}$ as the signature for the hottest surface temperature may introduce certain inaccuracy, since $\mathscr{T}$ measures the average temperature over the sample and the hottest surface temperature may occur at a time slightly earlier than that for this largest drop. A rough estimate shows that this can lower the calibrated temperature values by as much as 10 %. Hence, we conclude that within experimental error, the present measurements and calibrated results, the theoretical calculations, and the concept of strongly superheated liquid film at the interface, are all consistent with each other.

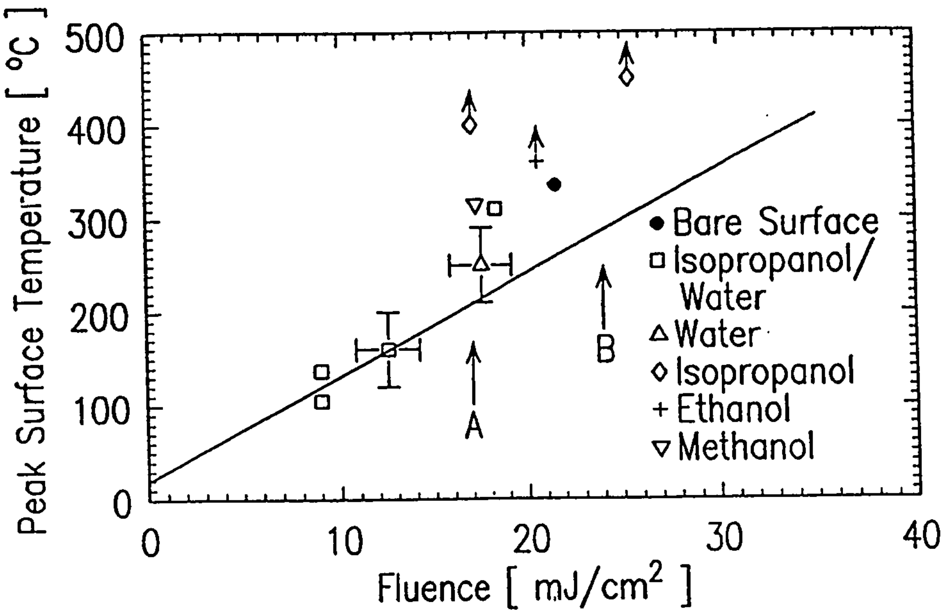

Figure 3.9(a): Comparison between calibrated results with theoretical calculation for the 0.2 μm a-Si sample. The arrows are shown for the threshold as in Fig. 3.4(a). The peak temperature data derived for isopropanol and ethanol go beyond the scale.

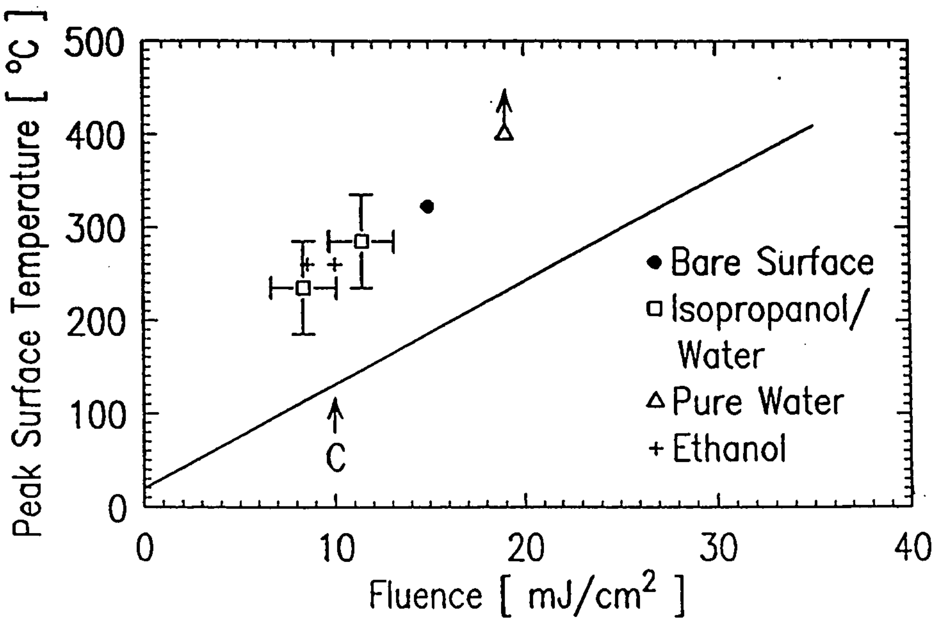

Figure 3.9(b): Comparison between calibrated results with theoretical calculation for the 1.0 μm a-Si sample. The arrow is shown for the threshold as in Fig. 3.4(b). The peak temperature derived for pure water goes beyond the scale.

3.5. Experimental Conclusion

Through the application of an optical transmission technique, we have been able to monitor the temperature and nucleation dynamics of an explosively superheated liquid film in the vicinity of an opaque substrate. Results obtained support the picture for a strongly superheated thin liquid layer at the interface upon the absorption of the excimer laser pulse ($\sim 10ns$) by the substrate. Huge transient liquid pressures are then generated at the interface, which provides a mechanism for particulate removal in the actual laser cleaning process [5 – 8]. We conclude that factors like the surface tension of the liquid are crucial, where others like the liquid film thickness are relatively insignificant in such film explosion arrangements. In addition, our approach provides further information concerning the nucleation dynamics of the interfacial liquid through the appearence of an extra dip in the transmission signal. This second dip reveals rich dynamics, including an analogy to critical opalescence, degree of heterogeneous nucleation, and possible supercritical phenomena. It is this last issue that we feel to be of great interest and worth further investigation. If the superheated liquid is really insulated from the substrate surface as we speculated in the above, then we have to think of some other method to probe its temperature. The technique of photothermal probe-beam deflection spectroscopy may be useful for this study and is being investigated.

Chapter 4

Conclusion

With the significant dependence of the optical constants of a-Si on temperature, we have been able to monitor the transient temperature at the liquid-solid interface and to observe the dynamics of the laser-induced superheated liquid films. In the study of the temperature dependence of the optical properties for a-Si, the most remarkable findings are the obvious temperature dependence of the refractive index, contrary to those previously reported [1 – 3,4], and that of the optical band gap deduced by using the Mott-Davis formula, although this has not been used in the calibration of the transient temperature. We also noticed that the temperature dependence varies for different sample thickness, probably due to the condition of preparation. However, we expect that the qualitative behavior for such physical quantities should have some general validity.

The variation of the optical constants for a-Si with temperature has enabled us to go on monitoring the transient temperature at the liquid-solid interface of a liquid/a-Si structure during the short laser pulse. Although the results are in general agreement with the published data in literature, many uncertainties and sources of errors must be considered. First of all, the approximate triangular shape with a halfwidth of 16 ns of the UV laser pulse used in the calibration is not always the real shape. However, we are assured [5] that this does not contribute any significant uncertainty in laser energy. Secondly, in fitting n and k, we have also introduced an inaccuracy into the calibrated temperature profile. In addition to the uncertainties coming from the laser fluence, small signal to noise ratio at low fluences, oscilloscope reading, we should remark that our identification of the deepest drop in $\mathcal{T}$ as the signature for the hottest surface temperature may introduce certain inaccuracy, since $\mathcal{T}$ measures the average temperature over the sample and the hottest surface temperature may occur at a time slightly earlier than that for this largest drop in transmission.

Other aspects of the mechanism of laser ablation of thin liquid films on opaque surfaces include the explosive pressure of the liquid, the force exerting on the contaminated particulates, and the trajectory of the particles. Some preliminary results for the explosvie pressure of the liquid have already been observed.

In conclusion, this is an experimental study of the superheating of liquids on a-Si thin film caused by a short ultraviolet excimer laser pulse. This establishes a primary understanding of the mechanism of the laser ablation of thin liquid films on opaque surfaces and should stimulate a deeper theoretical understanding of the mechanism.

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Chapter 4

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