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Time-Resolved Electron Thermal Conduction by Probing of Plasma Formation in Transparent Solids with High Power Subpicosecond Laser Pulses

**B.-T. V. Vu
(Ph.D. Thesis)**

Manuscript date: February 1994

LAWRENCE LIVERMORE NATIONAL LABORATORY
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MASTER

**Time-Resolved Electron Thermal Conduction by Probing of
Plasma Formation in Transparent Solids with High
Power Subpicosecond Laser Pulses**

by

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B.S. (University of Northern Iowa, Iowa) 1986

M.S. (University of California, Davis) 1988

DISSERTATION

Submitted in partial satisfaction of the requirement for the degree of

DOCTOR OF PHILOSOPHY

in

ENGINEERING - APPLIED SCIENCE

in the

OFFICE OF GRADUATE STUDIES

of the

UNIVERSITY OF CALIFORNIA

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1993

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1993

Time-Resolved Electron Thermal Conduction by Probing of Plasma Formation in Transparent Solids with High Power Subpicosecond Laser Pulses

by

BRIAN-TINH V. VU

Abstract

This dissertation work includes a series of experimental measurements in a search for better understanding of high temperature (10^4 - 10^6 K) and high density plasmas (10^{22} - 10^{24} cm $^{-3}$) produced by irradiating a transparent solid target with high intensity (10^{13} - 10^{15} W/cm ^{2}) and subpicosecond (10^{-12} - 10^{-13} s) laser pulses. Experimentally, pump and probe schemes with both frontside (vacuum-plasma side) and backside (plasma-bulk material side) probes are used to excite and interrogate or probe the plasma evolution, thereby providing useful insights into the plasma formation mechanisms. A series of different experiments has been carried out so as to characterize plasma parameters and the importance of various nonlinear processes. These lead to better understanding of the roles, competition and interplay between the processes, allowing one to effectively isolate electron thermal transport from other mechanisms that are of less importance. Experimental evidence shows that electron thermal conduction is supersonic in a time scale of the first picosecond after laser irradiation, so fast that it was often left unresolved in the past.

The experimental results from frontside probing demonstrate that upon irradiation with a strong (pump) laser pulse, a thin high temperature (~ 40 eV) super-critical density

($\sim 10^{23}/\text{cm}^3$) plasma layer is quickly formed at the target surface which in turn becomes strongly reflective and prevents further transmission of the remainder of the laser pulse. In the bulk region behind the surface, it is also found that a large sub-critical ($\sim 10^{18}/\text{cm}^3$) plasma is produced by inverse Bremsstrahlung absorption and collisional ionization. The bulk underdense plasma is evidenced by large absorption of the backside probe light. A simple and analytical model, modified from the avalanche model, for plasma evolution in transparent materials is proposed to explain the experimental results. Elimination of the bulk plasma is then experimentally illustrated by using targets overcoated with highly absorptive films.

Time-resolved study of electron thermal conduction is performed with coated targets where a supersonic thermal wave is generated by electron thermal transport and propagates into the bulk region. Information about the mechanism is inferred from time-resolved measurements of reflection, transmission and frequency shifts of the backside probe light. Calculations from the nonlinear heat wave model are shown to be in good agreement with the observations, suggesting that electron thermal conduction is the most effective energy transport mechanism during the first picosecond after laser irradiation where plasma expansion and other competing nonlinear processes are negligible.

In addition, when the laser intensity is kept below the plasma formation threshold, but still sufficiently high to modulate the refractive index of the transparent solid, time-resolved induced phase modulation effects in counter-propagating pulses can be observed by recording the frequency shifts in the backside probe light. The results, together with a simple model, offer a new technique of measuring temporal profiles of the ultrashort laser pulses. The technique determines the timing between the pump and probe pulses within 20% of the pulsewidth, complementing the timing of the plasma production and dynamics.

This research is comprised of:

1. Construction of the pump and probe experiments with frontside and backside probe beams. The experiments are so designed as to encompass the totality of the plasma evolution and render complementary information about the plasma formation and important energy transport mechanisms. Also, time-resolved amplitude (reflectivity and transmissivity) and phase (frequency shift) measurements of the probe pulse are simultaneously monitored to provide a complete basis for characterization of the laser light interacting with the plasma.

2. Experimental observations of induced phase modulation effects in transparent media in a nonlinear regime to obtain intensity thresholds for competing nonlinear effects and additional timing information about the plasma formation process.

3. Time-resolved measurements of reflectivity, transmissivity and frequency shifts using front (vacuum-plasma side) probe to confirm and reveal characteristics of the overdense surface plasma and its subsequent hydrodynamic expansion. In addition, time-resolved measurements of reflectivity, transmissivity and frequency shifts using back (plasma-solid side) probe provide direct observations of the ionized region behind the surface and thus laser-plasma coupling and the important energy transport mechanisms.

4. Energy dependent measurements of energies and spectra of the high intensity pump pulse transmitted through transparent targets make available further evidence of strong ionization or plasma effects, self phase modulation effects, and pulse shortening effects. Also, timing and intensity thresholds of these important effects are estimated.

Dedicated to

my late father

Can V. Vu

for his teaching and insurmountable sacrifice

and to my mother

Dinh T. Le

for her unconditional love and tender care

and to

all my brothers & sisters

for their encouragement and inspiration

ACKNOWLEDGMENTS

Here I would like to take this opportunity to offer my most sincere appreciation to a group of special people without whose support and influence this thesis project would have not been possible. Special thanks are due to:

Dr. Robert Miller & Mrs. Rachel Miller of Waterloo, Iowa, for their help during the very beginning of my journey in The United States as I try well to learn and speak "Good morning, America . . ." to my new friends.

Dr. Abraham (Abe) Szoke, my thesis research advisor, of the Lawrence Livermore National Laboratory for many things. Also, as my mentor Abe often goes out of his ways to offer help for which I am deeply grateful. His constant support and enthusiasm throughout the years of my research have helped me to overcome many obstacles, including scientific and technical problems. And most important of all, I thank Abe for having strong confidence in my effort and performance.

Professor Neville C. Luhmann, my thesis committee chair-person, of Dept. of Applied Science, University of California at Davis (U.C.D.) for his kindness and many suggestions to the style of the thesis.

I would like to acknowledge the other member of my thesis committee, Professor Ronald Soohoo of Dept. of Electrical Engineering, U.C.D, for many informative discussions of science and academe and his kindness.

Dr. Otto (Nino) L. Landen of LLNL for all of his help and valuable advice in making this project possible. Nino is also very much credited for teaching me everything

about laser-plasma experimentation, and for many helpful discussions of important physics and many valuable suggestions in the final analysis of the experimental results for publications.

Professor Roger Haas of Dept. of Applied Science, U.C.D., for many illuminating discussions of physics and useful advice on the research process. Also, I would like to thank Prof. Haas for introducing me to Dr. Szoke, whose interest has brought my attention to this work.

Dr. Andrew U. Hazi and Dr. Rudolf W. Bauer of LLNL for their generosity and encouragement in supporting most of the capitals and resources much needed for the success of the project.

Dr. Mark J. Eckart and Dr. Richard E. Stewart of LLNL for allowing access to the dye femtosecond pulse laser and the experimental equipment in the Ultrashort Pulse Laser facility at LLNL.

Dr. Michael E. Campbell of LLNL for providing some experimental apparatus and use of the dye sub-picosecond laser in the Short Pulse Laser facility at LLNL.

The physicists at Ultrashort Pulse Laser facility for their help when technical difficulties often arise. Especially, Dr. L. Van Woerkom and Dr. W. White for teaching me to operate the complex femtosecond laser system, and D. Price for useful technical advice.

William Cook of LLNL for the technical advice during the design and construction of the target holder and translator for the experiments.

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Preface

Recent developments in the generation of subpicosecond laser pulses have offered new possibilities to reveal ultrafast phenomena that were key processes unresolved in the past. Such short laser pulses have been utilized to uncover various nonlinear effects. For example, the studies of self-phase modulation and induced frequency shifts that are associated with the nonlinearity in the refractive index of nonlinear media, n_2 ,¹⁻³ promise many new applications in future communication systems.⁴ Time-resolved spectroscopy with ultrashort pulses allows one to study nonequilibrium electronic states of condensed matter.⁵⁻⁷ In particular, subpicosecond laser pulses with sufficient energy ($\sim 1\text{mJ/pulse}$) have made possible generation of ultrashort lived ($\sim 10^{-12}\text{s}$), high temperature ($\sim 10^4$ - 10^6K) and high electron density plasmas (10^{22} - $10^{24}/\text{cm}^3$) that are of current interest for generation of ultrafast x-ray sources.^{8,9} Until recently, the important physical mechanisms involved on the subpicosecond time scale and characterization of such plasmas remain unexplored, and the present availability of ultrashort (10^{-12} - 10^{-13}s) pulse lasers now allows one to commence such studies of the plasmas.

When focusing laser pulses of subpicosecond duration onto a solid target, absorption of laser radiation, hence electron heating, rapid ionization and collisional excitations occur within the penetration depth ($\sim 100\text{\AA}$) of the laser light. This leads to formation of a high temperature layer of plasma, composed of free electrons and ionized particles, at density of solids. As the plasma rapidly cools off, it emits x-ray radiation by line emission, recombination and Bremsstrahlung mechanism.⁹ The rapid cooling of the plasma is due to radiative processes, hydrodynamic expansion¹⁰ and fast conduction of thermal electrons¹¹⁻¹³ into the underlying cold region behind the target surface.

Clearly, the creation of such a plasma is intrinsically violent, and the plasma itself is extremely chaotic and undergoes many interacting channels of nonlinear processes that are seemingly impossible to comprehend. This henceforth poses a formidable task for both experimental and theoretical researchers to seek a quantitative and tractable description of the laser-produced plasmas, a challenge as pure as one described by Lord Kelvin :

I often say that when you measure what you are speaking about, and can express it in numbers, you know something about it, but when you cannot express it in numbers, your knowledge is of a meagre and unsatisfactory kind; it may be the beginning of knowledge, but you have scarcely, in your thoughts, advanced to the stage of science, whatever the matter may be.

On the time scale ($>10^{-12}$ sec), where motion of particles or nonlinear processes such as hydrodynamic expansion and atomic processes such as recombination are significant, the problems to characterize the laser-produced plasma appear indeed intractably difficult by virtue of the roles and interplay between many atomic and nonlinear processes. However, as we look for novel ways such as pump-probe techniques and new tools such as femtosecond lasers for better insights and directions, we often come to realize in a different regime where new order out of chaos may be found as inspired in the words of Y. R. Shen:¹⁴

... In generating and heating a plasma, the laser irradiation creates such chaos that all imaginable processes could take place. Yet the history of science has witnessed many such successes. Through perseverance and intelligence, one can always hope to find order in chaos. It is in fact the creation of order out of disorder that brings joy and excitement to devoted scientists.

There lies the very philosophical foundation for my experimental objective. My goal is to investigate the important mechanisms responsible for plasma production in transparent materials and to characterize plasma evolution both in time and space. Experimental approach to this goal is that a series of different experiments is carried out, so as to quantify the importance of various nonlinear processes. These experiments reveal different difficulties involved in efforts to separate electron thermal transport from other competing processes. In addition, theoretical models are proposed to account for propagation and absorption of high intensity ultrashort laser pulses, and thereby important plasma parameters. Experiments are planned and carried out to obtain time-resolved amplitude (reflectivity and transmissivity) and phase (frequency shifts) measurements of the probe light interacting with the plasma. These results are then compared to calculations for inference of the dynamics of the plasma and the important mechanisms.

This dissertation is organized in the following manner : Chapter 1 presents an overview and requirements to pursue the pump and probe experiments and their advantages over conventional laser-produced plasma experiments. The experiment was initially carried out with a picosecond laser system, and then with a femtosecond laser as soon as it became available. Descriptions of both lasers, followed by a detailed description of the experimental configuration and schematics, are given in Chapter 2. In Chapter 3, I begin with the first experiments and studies on phase modulation effects induced by the strong pump pulse on the weak counter-propagating probe pulses. The results are then compared with computer simulation to determine the absolute timing between the pump and probe pulses that is useful for subsequent measurements on laser-produced plasmas. The second part of the research project is given in Chapter 4 where evidence for formation of a super-critical plasma layer taking place at the target surface is confirmed. Also, the overdense surface plasma temperature and characteristic time

constants for its subsequent expansion are deduced from the observations. Evidence for, and characterization of, a large sub-critical plasma in the bulk region behind the surface together with a modified avalanche model for plasma evolution in the bulk materials are shown in Chapter 5, as the third part of the project. Illustrations to eliminate the bulk plasma by using targets overcoated with highly absorptive thin films are presented, indicative of formation of a highly reflective super-critical plasma layer by electron thermal conduction. Chapter 6 shows experimental measurements of frequency shifts in the backside probe light with coated targets where a supersonic thermal wave supported by electron conduction acts like a highly reflective moving mirror. Comparisons with the theoretical predictions indicate good agreement with the nonlinear heat wave model, confirming electron thermal conduction as the most dominant energy transport mechanisms during the first picosecond after the femtosecond laser irradiation. This, as suggested by the title of this thesis, is the central part of this research. Chapter 7 shows experimental data and calculations for characteristics of the propagation, absorption and temporal shortening of high intensity laser pump pulse transmitting in the transparent materials. Finally, in Chapter 8 I summarize the experimental findings and their important implications, and I outline future experiments along the line of these investigations. The appendices are added at the end to describe miscellaneous aspects of the experiments such as algebraic details of the analytical models, geometrical analysis for laser beam-target alignment, computer codes for simulation of the data and for driving target raster system, and a table of useful and often used formulae.

CHAPTER 1

MOTIVATION AND OVERVIEW OF THE EXPERIMENTS

The interest and motivation of this thesis research have grown out of many fundamental questions raised in the physics of inertial confinements fusion concepts and laser-plasma interactions. In recent years, utilization of high power ultrashort pulse lasers to produce for the first time high temperature plasmas of solid density¹⁵⁻¹⁷ has opened up new ways to study fundamental properties of extremely hot and dense matter that is heated too rapidly for any hydrodynamic processes.^{15,16} Such studies include physics of plasmas in a new and interesting regime of solid-state density,¹⁷ and physics of atoms subjected to strong laser fields.¹⁸⁻²⁰ As the prospects and physics of using x-ray emitting plasmas to pump x-ray lasers are being further investigated,^{15,21,22} many new methods and techniques have been developed to study ultrafast phenomena in various disciplines. For example, time-resolved x-ray diffraction studies of electronic structure of materials and biological systems, x-ray lithography and holography, and surface dislocation and crack formation in metals.^{23,24} It is therefore important to understand what the requirements of the materials and the laser sources for production and characterization of such plasmas are. Also, investigations of the important mechanisms must be made for efficient laser-matter coupling before any technological applications of the plasma becomes economically feasible. These considerations have led us to employ the techniques and diagnostics discussed below.

In the following, I will give an overview of my experiments, conditions and findings with picosecond and femtosecond pulse lasers for production of plasmas and important

mechanisms. The design of the experimental set-up is similar to the conventional pump-probe experiment, but with some technical modifications to obtain complementary information about different mechanisms involved in the plasma formation. As will be seen, the experimental design and diagnostics represent a complete pump-probe configuration to obtain time-resolved dynamics of the plasma evolution. The complete configuration consists of schemes for production of hot and dense plasmas, diagnostics for both front and back probing of the plasmas, and measurements of both amplitude and phase information of the probe light interacting with the plasmas. The frontside probing confirms formation of a high temperature (≈ 40 eV) and solid density ($\sim 10^{23}\text{cm}^{-3}$) plasma layer at the target surface, and determines the characteristic time scale of the plasma expansion into the surrounding vacuum. On the other hand, the backside probing, unaffected by the plasma expansion, directly interacts with the bulk region behind the surface whose changes in the material properties are a result of energy transport phenomena. I will point out the approaches and both advantages and disadvantages to tackle the complexity of the laser-induced plasmas and circumvent the difficulties as we come to realize at each step taken in the course of the different experiments. This leads us to acquire better understanding of various nonlinear processes and their implications. Consequently, successful separation of the fast electron thermal transport from other competing processes may be attained.

While high temperature and density plasmas are readily achieved with laser pulses of many picoseconds or longer, irradiation of solid transparent targets with high intensity (10^{12} - 10^{15}W/cm^2) ultrashort (10^{-13} - 10^{-12}sec) laser pulses however offers several advantages for production of hot plasma at solid density:

1. In heating with long pulses, hydrodynamic expansion of the plasma into the surrounding vacuum often takes place significantly prior to the end of the heating pulse, taking away the energy of electron and reducing the density of the plasma. It is thus necessary to utilize sufficiently short duration pulses so that laser energy deposition to the target occurs quickly before plasma expansion becomes appreciable. Also, probing of plasmas using short pulses provides higher temporal resolution measurements.

2. Solid materials, typically about 10^4 times denser than that of gases, by virtue of their rigid structure and essentially no density variation make easier to limit and fix a desired number of atoms exposed to the laser radiation. Thus, the plasma can be readily heated to a very high temperature at solid density.

3. If the laser field exceeds or is comparable to that of atomic Coulomb field, neutral atoms may be ionized by multiphoton and inverse Bremsstrahlung absorption; and bound electrons become loosened and heated easily under the influence of the laser field because of high collisionality at high density. High intensity laser light is thus required for efficient laser-matter coupling and production of high temperature plasmas.

4. Transparency of the target is required mainly for direct probing from the backside of the plasma in the bulk region into which the absorbed laser energy is transported via electron thermal conduction. In addition, transparent targets largely eliminate the effect of a laser pre-pulse that otherwise may be sufficient to pre-ionize and ablate the target surface prior to the arrival of the main pulse.^{25,26}

FIG.1.1: Laser beam-target interaction geometry

In irradiation of a transparent solid target with a high intensity laser (pump) pulse, as shown in Fig.1.1, a highly reflective super-critical plasma disk-like layer (diameter $d=75\mu\text{m}$) is formed at the surface (Chapter 4). This layer is strongly reflective and stops transmitting any laser light, and thus prevents further increase in the plasma production during the remainder of the pump pulse. It is this layer that limits the laser-plasma coupling and laser-to-x-ray conversion efficiency. The front side probe light, strongly reflected from the critical surface where the plasma frequency is equal to the laser frequency, cannot interact with the bulk region behind the surface. If the probe pulse arrives at the surface late relative to the heating pump pulse, the reflected probe light will experience large absorption as the plasma layer expands and becomes sub-critical, thus determining the temperature and the characteristic time constants of the expanding surface plasma.

In order to examine what really happens in the bulk region behind the critical surface and to gain more information about the laser-plasma coupling and transport mechanisms, the probe light is allowed to interact with the plasma from the back side as shown in

Fig.1.1. The backside probe beam is at 19° to the normal of the surface. This angle of incidence fixes the interaction length between the pump and the probe, $L=120\mu\text{m}$. The backside probe light is kept from interacting with the expanding plasma because of the overdense plasma layer, and the reflected backside probe light interacts directly with the bulk region. Changes in the material properties of the bulk region then can be extracted from the amplitude and phase changes in the reflected backside probe light. This is thus complementary to the frontside probing data. As will be seen later, in contrast to frontside probing where the probe reflection is enhanced, the backside reflected probe light suffers large absorption (Chapter 5). This is clear evidence for formation of a large sub-critical plasma in the bulk regions, in addition to a strongly reflective overdense plasma layer at the surface. Formation of the bulk underdense plasma is strongly regulated by inverse Bremsstrahlung absorption and collisional ionization of readily-ionized impurities in the target materials.

Because of the bulk underdense plasma and its large absorptivity, both backside probe transmission and reflection signals are strongly absorbed. These signals are too weak for any meaningful interpretation about production of the overdense surface plasma and transport mechanisms involved. The problem is then to eliminate the bulk plasma so that the reflected backside probe light signal is only affected by the overdense surface plasma. Elimination of the bulk plasma is experimentally illustrated with targets overcoated with a highly absorptive thin film (Chapters 5 & 6). The role of the thin film is to create an overdense plasma at much lower intensity level and earlier during the leading edge of the pump pulse. A large reflection increase in the probe light is observed, strongly indicating the evolution of a transient overdense plasma at the surface and the disappearance of an otherwise highly absorptive underdense plasma in the bulk region.

The reflection enhancement in the backside probe light with coated targets is found to have a risetime comparable to the heating pulse duration and a falltime of many picoseconds. The experimental results acquired up to this point suggest that electron thermal conduction is the main transport mechanism to account for such short risetime and long falltime in the reflection enhancement (Chapter 6). It is found that the overdense surface plasma layer is produced during the leading edge of the pump pulse.²⁷ As a result, the subsequently arriving portion of the pump pulse is kept from further interacting with the region behind the critical surface. It however continues to deposit its energy in the surface region near the critical density, resulting in a large temperature gradient at the surface.¹² Because the ions in the plasma are much heavier than the electrons ($M_i/m_e \approx 4 \times 10^4$), it takes some time for plasma expansion or motion of the ions to initiate. As will be experimentally shown, this time is about 1ps, and the time for plasma expansion becomes significant is ≈ 6 ps, much longer than the duration of the pump pulse. Prior to appreciable plasma expansion or development of a shock wave that results from expansion, the absorbed energy is transported into the bulk region via conduction of electron from hot to colder regions. Thus, another role of the absorptive thin film is to set up a high temperature (≈ 40 eV) heat source at the target surface for subsequent generation and propagation of a thermal wave supported by electron thermal transport.^{13,28} As the thermal wave moves into the transparent materials, it thermally ionizes the neutral atoms, leading to a high temperature and super-critical density plasma layer. As predicted by the nonlinear heat wave model,¹³ which will be shown later, the thermal wave is initially supersonic and eventually become subsonic due to the increasing volume of the plasma.

To further confirm the dominance of the electron thermal conduction and test the validity of the nonlinear heat wave model experimentally, we record time-resolved frequency shifts in the backside reflected probe light prior to plasma expansion (Chapter

6). The measurements reveal supersonic motion of the ionization front that acts like a highly reflecting moving mirror during the first picosecond after laser irradiation. These shifts yield an estimate of $1.8 \times 10^7 \text{ cm/s}$ for the maximum velocity of the ionization front at the critical density. This is six times greater than the maximum ion acoustic or sound velocity of the plasma ($3 \times 10^6 \text{ cm/s}$) that was measured by frontside probing of the plasma.

Additional information about the ionization rate is obtained by monitoring spectral changes in the high intensity (pump) light transmitting through the transparent target (Chapter 7). The spectral changes are recorded as a function of peak intensity of the incident pump pulse. They are used to infer ionization rate, characteristic time constants and various intensity thresholds. Further information on the timing of plasma evolution and dynamics can also be acquired by measuring phase change or frequent shifts in the counter-propagating probe pulses (Chapter 3). In this phase modulation experiment, the pump pulse intensity is attenuated, below the plasma formation intensity threshold, so that the probe light suffers a frequency shift which is linear proportional to the product of the nonlinear refractive index of the glass n_2 and the pump pulse intensity profile $I(t)$.

The results of this research are compared with the predictions made by the existing models. The measurements on the phase modulation effect observed in counter-propagating pulses show good agreement with the proposed model and consistent with the existing model (Chapter 3). Observation and confirmation of an overdense plasma layer (Chapter 4) are similar to the works of other researchers, despite the difference in the laser parameters, e.g. laser pulse length or energy. Evidence of the underdense plasma in bulk targets (Chapter 5) is observed for the first time; and the experimental measurements are in excellent agreement with the modified avalanche model that is also presented in this thesis. Experimental evidence about the supersonic thermal wave driven into the solid target by 100fsec laser pulses shows fairly good agreement with a nonlinear

heat wave model that is a combination of the Marshak's wave²⁹ and the Zeldovich's wave models.¹³

Implications of this research and its results are numerous. In laser fusion experiments, the density of the compressed materials in a target is limited by the maximum ablation pressure and preheating of the materials.^{30,31} The ablation pressure ($\sim n_e T_e$) is determined by the laser-plasma coupling efficiency in the absorption region at the target surface; whereas preheating of the region behind the surface is strongly influenced by x-ray photons emitted from the surface plasma and/or by the flux of the super-thermal electrons. Clearly, these are phenomena of thermal transport and laser-plasma couplings that critically determine both the plasma conditions and target implosion efficiency. The results of this research will allow us to test and develop fundamental understanding of several aspects of basic physics of high temperature and solid density plasmas. For example, the classical models, such as that of Debye-Huckel to account for screening effects of charged particles, or that of Saha-Boltzmann for conditions in a thermal equilibrium, successfully describe most low density or astrophysical plasmas. They however may be no longer valid for plasmas that are obtained by ultrashort high intensity laser pulses.¹⁹ Such plasmas are used as ultrashort x-ray emitting sources or as a gain medium in x-ray lasers. Understanding of these plasmas have been realized to be of great importance in the development of these applications. It is therefore essential to understand what regulates and limits laser absorption and hence conversion efficiency of x-rays emitted from the plasma. This is an intrinsic difficulty in the current development of short and intense x-ray sources that hold the promise of time-resolved structural and electronic studies of solids and short-lived chemical or biological systems. Characterization of the transport mechanisms and limiting factors will enable us to technically improve laser absorption and thus the technology of high power laser energy conversion. This is so since the brightness and dimensions of x-ray sources are

profoundly determined by the temperature-density structure in the plasma which in turn is strongly regulated by the thermal transport properties and laser-plasma coupling. Another application of this study, which is of no less importance in laser research, is implied in the experimental technique itself. The pump and backside probe technique illustrates a new way for the first time to time-resolve laser-induced surface and bulk damage phenomena that are critical in the technology of high power tolerant laser materials.

CHAPTER 2

EXPERIMENTAL APPARATUS & PLANS

I describe in this chapter the picosecond laser system that has been used for our first experiments at the time when laser technology was rapidly developing toward higher power and shorter pulse regime. In fact, a few months after the induced phase modulation experiments and time-resolved measurements of the plasma with the picosecond laser succeeded, high power femtosecond lasers became available for subsequent experiments on plasma studies. A description of the femtosecond laser used in subsequent experiments will also be given.

The picosecond laser together with the gain quenching device produces pulses with no post-pulse, of duration ranging from 0.7 to 1.2ps and energy of about 1-2mJ per pulse. The femtosecond laser system produces pulses of length varying between 100 and 200fs and energy of about 1-2mJ per pulse. Because of shorter pulse length, the femtosecond laser delivers higher power by a factor of about 10, making the laser-produced plasma hotter and denser and allowing better time resolution measurements.

The configuration of the pump-probe experimental set-up also will be described in detail. The set-up was designed with both frontside and backside probing beams for amplitude (reflectivity and transmissivity) and phase (frequency shifts) measurements. In addition, optical diagnostics for energy and spectral measurements of the high intensity transmitted pump light are included. The configuration presents a state of the art relatively complete optical probing measurement of the plasma evolution, though by no means revealing a complete picture of the laser-plasma interaction.

2.1 High Power Picosecond Laser & Post-Pulse Suppression Technique

The early design of the picosecond laser system built by workers at the Lawrence Livermore National Laboratory includes both commercial components and customized components. A detailed schematic and configuration of the laser has been given by Perry.³² The whole system is an amplified synchronously-pumped mode-locked dye laser. It consists of a commercial CW mode-locked Nd: YAG oscillator, linear dye laser, regenerative amplifier and a series of three dye amplifier cells. The system begins with the CW mode-locked Nd:YAG oscillator operating at 125MHz and producing 100ps pulses at 1.06 μ m with average power between 70-100W. These pulses go through a type II second harmonic (SH) KTP crystal to convert 1.06 μ m fundamental light into pulses at 0.53 μ m. The SH light is separated from the fundamental light by a dichroic plate and then directed to synchronously pump a commercial linear-cavity dye laser.

In the linear cavity, a Rhodamine 6G solution of $\approx 1 \times 10^{-4}$ M concentration is used to produce 1.5ps laser pulses of ≈ 2 nJ with high stability. Large tunability between 570-620nm is achieved by carefully adjusting the cavity length equal to the cavity length of the pump Nd:YAG laser. When the dye laser cavity length is made slightly shorter than that of the pump laser, pulses as short as 0.6ps can be obtained, but accompanied by satellite post-pulses which may not be suitable for the study of laser-produced plasmas.^{26,33} As will be shown later, to ensure no post-pulses a optics scheme is built at the output of the linear dye laser.

The dye amplifier consists of three cells flowing with Sulfurhodamine B solutions of concentrations 2, 1 and 0.8×10^{-5} M. Variable delay lines for each cell are used to maintain precise timing of the pump laser pulse and the dye laser pulse from the linear dye laser. Precise timing of the pump and dye laser pulses arriving at the cells are

important for reducing the amplified spontaneous emitted radiation. The final output pulse is about 1-2mJ per pulse with a pulselength of 0.7-1.2ps. The shortest output of 300fs can be achieved by fine tuning the linear cavity length, but it is highly unstable and accompanied with satellite post-pulses.

The critical part of the laser is to minimize ASE light and to suppress the pre- and post-pulse levels. Fine tuning of the dye laser cavity length and short-pulse pumping of the dye amplifiers reduces the pre-pulse level.³⁴ During experiments the fluence of the pre-pulse accompanying the main pulse is always kept below 0.1J/cm² to prevent any plasma formed before arrival of the main pulse. The main pulse is always accompanied by a post-pulse of about 4ps long, and 7-8ps after the main pulse. Energy of the post pulse accounts for about 10% of the total energy. Post-pulse elimination has been accomplished by a gain-quenching technique as described in detail by Landen and Szoke.³⁵

FIG.2.1: Post-pulse suppression set-up

Figure 2.1 shows the gain-quenching scheme for the post-pulse suppression set-up. The set-up is implemented near and prior to the first dye amplifier cell. A 50% beam splitter is used to divide the unamplified dye (seed) pulse into two pulses, one as the main pulse (B) and the other as an independent gain-sweeping pulse (A1, A2). The former goes through a variable delay line to compensate for the round trip of the latter to obtain precise timing. Figure 2.2 shows in greater detail the precise timing of the pump pulses (C1, C2), the gain-sweeping pulse (A1, A2) and the desired main pulse (B). The gain-quenching pulse is first amplified to sufficiently high energy (A1) by the first amplifier. This gain sweeping pulse (A2) then reenters the gain cell about 3-4ps after the main pulse (B) as shown in Figure 2.2. Effectively, this maintains energy gain seen by the main pulse (B), and at the same time it reduces the amplification factor that would otherwise be enjoyed by the post-pulse of the main pulse. Thus, the contrast between the main pulse and post-pulse is profoundly enhanced and can be optimized by fine tuning the time delay ($\sim 1\text{mm}$) and using a variable beam splitter to slightly vary the input energy of the gain-sweeping pulse. If the energy of the gain-sweeping pulse is too low, it cannot suppress the energy gain available after the main pulse and before the postpulse. On the other hand, if it is too high, the gain sweeping will be complete, but energy of the main pulse will be reduced. Optimization of the contrast depends on several factors, i.e. available fluence, gain-length product, and is sensitive to the beam splitter reflectivity and timing between the pulses.

FIG.2.2: Timing diagram of the pump pulses, gain-sweeping pulse and main pulses

Figures 2.3 and 2.4 display a measured autocorrelator trace (dots) and a spectrum of the laser pulse obtained with the post-pulse suppression scheme. The trace is measured with a commercial single shot autocorrelator. The spectrum is an average of 150 laser shots, measured by a (33mm) spectrometer interfaced with an optical multichannel analyzer. The intensity levels at the peak and the wings of the laser pulse are examined by using a set of various neutral density filters to attenuate the laser energy and a 2nd order autocorrelator. Low density filters are used to bring out the shape and level of the pulse wings.³⁵ The contrast between the main pulse and the wing levels is found to be about 10^{-4} . The laser beam when focused to produce a plasma of 75 μ m in diameter has an intensity level of $\approx 2 \times 10^{13}$ W/cm² at the peak, and $\approx 10^9$ W/cm² level at the wing, much below the damage threshold ($\sim 10^{12}$ W/cm²). The measured autocorrelation trace is best fit to the normalized second order autocorrelation function (dashed curve) calculated by,³⁶ assuming a Sech² temporal shape with full width half maximum (FWHM) of 1.0ps (solid curve):

$$G^{(2)}(\tau) = \frac{\int_{-\infty}^{+\infty} I(t)I(t+\tau)dt}{\int_{-\infty}^{+\infty} I^2(t)dt} \quad (2.1)$$

where FWHMs of the autocorrelation function $G^{(2)}$ and Sech² are related by the relation:³⁷

$$\Delta\tau = 1.55 \Delta t \quad (2.2)$$

The spectrum of the laser pulse averaged over about 150 pulses shows a spectrum of 5 $\text{\AA}$ FWHM sitting on a weaker 13 $\text{\AA}$ wide component centered about the central wavelength at 580 $\text{\AA}$. Shot -to-shot spectral jitter is less than 0.3 $\text{\AA}$.

Figure 2.5 shows the comparison between the Sech^2 and Gaussian pulse shapes, indicating very little or essentially no difference in the pulse profiles. Since there is very little difference in the amplitudes far at the wings of the two shapes, effects of pulse shapes on the plasma formation are insignificant, and the use of either function is merely a matter of computational convenience

FIG.2.3: Measured autocorrelation trace of the laser pulse intensity (closed circles), normalized to its peak value. A laser pulse with a 1.0ps FWHM and Sech^2 shape is assumed and yields the best fit autocorrelation (dashed curve), in least square sense, to the measured trace.

FIG.2.4: Spectrum averaged from 150 laser pulses.

FIG.2.5: Calculated autocorrelation traces of picosecond laser pulses using Sech^2 and Gaussian pulse shapes with 1ps FWHM, indicating no significant difference between the two shapes.

2.2 High Power Femtosecond Laser

The femtosecond laser system was built by W. White et al.³⁸ at the Lawrence Livermore National laboratory. The system consists of a colliding pulse mode-locked (CPM) ring dye laser, a sequence of three customized dye amplification cells and a pulse compressor. Figure 2.6 shows the overall configuration of the system. Pulses from the CPM laser are amplified by a bow tie amplifier and two Bethune cells, in sequence, to produce a final output of 1-2mJ, 100-200fsec pulses with a repetition rate of 10Hz at a wavelength of 616nm.

FIG. 2.6 Overall configuration of the femtosecond laser system.

FIG.2.7 Configuration of a CPM laser

The colliding pulse mode-locked dye ring (CPM) laser was first designed by Fork et al. in 1981,³⁹ and later its modification and improvements have been demonstrated by Valdmanis et al.⁴⁰ Owing to its easy operation and its advantages in pulse stability and maintenance, the CPM laser has been commercialized and used widely in most present university laser laboratories. The laser, operating on the principle of passive mode locking as shown in Fig. 2.7, supports two oppositely traveling pulses and is pumped by an argon ion laser beam of 2.5-4W at the gain jet which contains Rhodamine 6G $5 \times 10^{-3} \text{M}$ and ethylene glycol solution. The two pulses collide at the $2 \times 10^{-4} \text{M}$ DODCI saturable absorber jet at which the combined energy of the two colliding pulses produces a deeper saturation of the absorber, thus lower energy loss. The pulses pass the absorber jet with reduced attenuation at the peaks and strong absorption at the wings of the pulses, hence the pulse FWHM is effectively shortened. Furthermore, in a medium with normal or positive group velocity dispersion (GVD), the red frequency component in the large bandwidth of the pulse travels faster than the orange or blue component, making the pulse temporally broadened. This broadening is then compensated by an amount of negative GVD of the prism glass.^{40,41} Thus, operation of the laser relies on the balance of the gain dispersion and gain saturation that are strongly determined by the gain and absorber

concentrations. Also, the output pulselength and stability critically depend on the compensation of the self phase modulation produced in the jets and reflecting mirrors which give rise to a positive chirp and the negative GVD determined by the series of four prisms. The total dispersion can be varied by adjusting one or two prisms, without deflecting the laser beam, to either increase or reduce the amount of the prism glass in the laser cavity.⁴² Because of its operation based on the principle of mode locking, the CPM spectral tunability is rather limited, only from 614 to 620nm, much narrower than that of the synchronized-pump picosecond laser. The real reason for this limitation is that so far no saturable materials with optical quality as good as DODCI has been found.

FIG.2.8: Measured autocorrelation traces from CPM laser (open circles) and amplified pulse output (close circles) are best fit to the calculated second-order autocorrelation trace (curve) given by Eqn.(2.1), assuming Sech^2 pulseshape with 120fs FWHM.

FIG.2.9: Spectrum of CPM output

Figure 2.8 displays the second-order autocorrelation trace measured from the CPM output, and its spectrum is shown in Fig. 2.9. The CPM output consists of two beams that are resulted from clockwise and counter-clockwise intracavity beams. One beam is used for real time diagnostics and continuous monitoring of the laser, including pulse duration, spectrum, and generation of triggering signals for subsequent amplification stages. The other CPM beam with 0.2nJ per pulse at a repetition rate of 90MHz is used as the main beam. It is the input for amplification in the bowtie cell.

The bowtie amplifier shown in Fig.2.10 uses six passes through a single gain medium to elevate the CPM pulses from subnanjoule to microjoule level per pulse. The gain medium, pumped by the residual light from Nd:YAG laser, was originally designed and demonstrated by Knox et al.⁴³ and Nickel et al.⁴⁴ After four passes through the gain medium, the beam is directed through a saturable absorber to suppress amplified spontaneous emission (ASE) down below ~1% of the main pulse. The bowtie output,

about 2-6 μ J per pulse, is then spatially filtered by a pinhole (80 μ m) to maintain high transverse mode quality and reduce ASE level before it enters the second amplifier.

FIG. 2.10: Schematic of the bowtie amplifier with saturable absorber.

The second and the last amplifier are both Bethune-type dye cells⁴⁵ of 6mm and 20mm in diameter, respectively. Figure 2.11 shows the end view of a typical Bethune cell. It is a prism with a cylindrical flow channel bore slightly off center. When the pump

light enters from the left, due to total reflections at the right-angle surfaces, the cylindrical column of dye solution is illuminated uniformly. Segments of the large pump beam, numbered 1, 2, 3, and 4, represent the images in the pump beam where the top, back, front and bottom of the dye column are pumped, respectively. For maximum pump efficiency, the prism surface facing the pump is anti-reflection coated and the right-angle surfaces are coated with highly reflective films. Because the dye cylindrical column is pumped from the four directions, uniform pumping and thus relatively good beam quality in the amplified pulse can be obtained with little alignment sensitivity to the pump beam.

Following the 20mm Bethune cell amplifier is a pair of gratings that are used to recompress the amplified pulse. The pulse at this point has traveled through many optical components (reflecting mirrors, glass cells), and it experiences positive group velocity dispersion (GVD), making the pulse with large bandwidth significantly lengthened in time. The broadening in the amplified pulse is then corrected by using a pair of parallel gratings which exhibit negative GVD.^{42,46,47}

FIG.2.11: Bethune prism-like dye cell.

FIG.2.12: Output beam radial intensity distribution at focal spot ($89\mu\text{m}$) in log scale.

The final output that is used for plasma study, after a pinhole (Fig.2.6) for spatially filtering of the beam, is about 1-2mJ per pulse. Pulse duration can be made stabilized and varying between 100-200fs at the center wavelength 616nm by fine tuning one of the prism in the CPM cavity. Variation of the pulse duration and energy is about 20%. Figure 2.12 shows the transverse profile of the laser $I(r)$ in log-scale at the focal spot after the third spatial filter, indicating a relative good focusability and mode of the output beam.

2.3 Experimental Set-Up and Details

Overall configuration of the experimental setup is shown in Fig.2.13. A transparent solid target is housed inside a vacuum chamber with pressure maintained at $\approx 10^{-5}$ - 10^{-4} Torr. The experiment begins with a laser pulse, going through a 99% beamsplitter and splitting into a stronger pump pulse and a weaker pulse. The strong pulse is normally incident at the target to produce a plasma on a surface. The weaker pulse is used to probe the plasma from the other side or the backside of the plasma, thus the name backside probing. The energies of the incidence, reflection and transmission of the backside probe

light (Reference, R_b and T_b) are simultaneously monitored by three (PIN 8LC) photodiodes (PD1, PD3, PD2, respectively). The signals from the diodes are preamplified and sent to gated boxcar integrators (Stanford Res. SR250) interfaced with a computer for data acquisition and reduction. The raster system is to translate the target so that each time the laser pulse arrives at the surface it will hit a fresh part of the target, maintaining the condition of plasma formation on each laser shot. Figure 2.14 shows the details of the actual design of the target holder, focusing lenses, diodes and horizontal and vertical tilt micrometers for target alignment. As the target is rastered, the delay line is moved in order to vary the relative time of the pump and probe pulses arriving at the plasma (front surface) and to allow time-resolved measurements. Spectral change and frequency shifts in the backside reflected probe light are also examined by a spectrometer (33cm, 1200 grooves/mm grating) interfaced with a high resolution ($0.3\text{\AA}/\text{pixel}$) optical multi-channel analyzer (OMA).

The mirror M2 is rotatable, so that the weaker beam can be redirected to probe the plasma on the same side with the pump or the frontside of the plasma, thus the name frontside probing. Again, the energies of the incidence, reflection and transmission (Reference, R_f and T_f) of the frontside probe light are monitored by the diodes (PD1, PD2, PD3, respectively) and are fed into boxcars. Spectral changes in the frontside probe reflected light are also recorded by the spectrometer. The lens L6 collects the high intensity light of the transmitted portion of the pump pulse for measurements of transmitted energy and frequency shifts, giving more detailed observations of the self phase modulation effects due to plasma formation.

FIG.2.14 Actual design of the target holder inside the vacuum chamber

The target is either commercial 1mm thick glass⁴⁸ or 1.6mm thick pure graded fused quartz slide.⁴⁹ Both materials are silicon dioxides, but the glass contains about 27% impurities of readily ionized materials. The target is so thick that interference of the reflected and transmitted probe light is prevented in the bulk region. At the rear surface of the target, there is an anti-reflection coating to suppress probe reflection from that surface (see Fig.1.1). Polarization of the pump beam is P-polarized, and the probe beam after passing through the halfwave plate is made S-polarized, so that stray diffuse light of the pump can be rejected before detection by the polarizers P located at the diodes. Furthermore, the analysis of the probe light will be simplified since the S-polarized probe light has no electric field component parallel to the plasma density gradient, and thus there is no resonance absorption.^{50,51} The focusing spherical lenses are 25cm and 7.5cm

focal length for the pump and the probe beam, respectively. The probe is tightly focused to a spot of $25\mu\text{m}$ diameter and centered on the larger focused pump spot of $75\mu\text{m}$ diameter, resulting a peak spatially averaged intensity up to a maximum of $2.0 \times 10^{13} \text{W/cm}^2$ with the picosecond laser and $5.0 \times 10^{14} \text{W/cm}^2$ with the femtosecond laser. Centering of the probe focal spot onto that of the pump is done as follows: A crater is made on the front surface with the pump. The probe is then tightly focused at the crater and either the reflection or transmission of the probe light is viewed on a screen. Figure 2.15 shows a typical crater of $\approx 75\mu\text{m}$ in diameter made on the surface as a result of a single laser shot incident on the target. The incidence angle of the probe is 30° to the normal at the rear surface corresponding to an internal angle of 19° at the front surface. From Fig.1.1 in Chapter 1 this laser beam-target geometry fixes the length of the interaction region between the pump beam and the backside probe beam between $100\text{--}120\mu\text{m}$, much smaller than the Rayleigh ranges of the pump (7mm) and probe (0.8mm) beams. The probe intensity is kept at the level of $\approx 2.0 \times 10^{10} \text{W/cm}^2$. This intensity is sufficiently low to be non-intrusive. It is below the threshold for growth rates of nonlinear processes, such as stimulated Raman or Brillouin scatterings,^{52,53} but still sufficiently high to discriminate the probe pulse specular reflection from the diffusely scattered light of the intense pump pulse. Timing of the probe relative to the pump is varied by a computer-driven delay line with $1\mu\text{m}$ accuracy in increments equivalent to few femtoseconds between laser shots. Velocity nonlinearity and other inaccuracies in the translation stage are all negligible compared to the spatial extent of the laser pulse ($\approx 300\mu\text{m}$). Spatial walkoff between the pump and probe beams at the target surface is kept below an acceptable $20\mu\text{m}$ throughout a temporal scan as follows. The horizontal and vertical tilt of the target are adjusted until the probe beam tracks the crater throughout a $5 \times 5 \text{cm}^2$ raster. Spatial alignment to maintain overlapping conditions of the two beams with $<10\mu\text{m}$ accuracy can be attained by iterating the geometrical algorithm that is discussed in Appendix D. The walkoff, translation stage wobble, and timing errors

(<100fs) due to target thickness variations are all negligible, thus ensuring the same conditions of plasma formation and probing on each laser shot.

FIG.2.15: Image of a crater produced as a result of single laser shot on the target

Rough absolute timing to within 20ps between the probe and the pump pulses is obtained with a fast photodiode (Hamamatsu R1328U-02). To determine the zero timing between the two initially counter-propagating pulses with better accuracy, we have performed induced phase modulation measurements.³ In these spectral measurements, the pump intensity is reduced to $\approx 1 \times 10^{12} \text{ W/cm}^2$. This intensity is below the plasma formation threshold, but still sufficiently strong to induce a modulation in the refractive index of the glass medium. The modulated index is given by $n_g(t) = n_{og} + n_{2g}I(t)$, where $n_{og}=1.52$ and $n_{2g}=0.9 \times 10^{-13} \text{ esu}$ are the linear and nonlinear refractive index, respectively, and $I(t)$ is the instantaneous intensity of the pump pulse profile. The spectral shifts of the reflected probe light are then resolved by a spectrometer interfaced with an optical multichannel analyzer. The frequency shifts for the return journey of the reflected

probe light are seen to be proportional to the time derivative of the pump intensity. At early delay time, or during the rising edge of the pump pulse, the probe suffers a red shift. At late delay time or during the falling edge, the probe experiences a blue shift. When the two pulses are perfectly overlapped in time at the surface, defined as the absolute zero delay time, there is no spectral shift observed in the probe light. The above is generally true for the probe pulse co-propagating with the pump pulse.⁵⁴ However, during its arrival to the front surface, the counter-propagating probe experiences an additional shift that is proportional to the accumulation of phase change as it traverses the pump pulse profile. Analysis of the spectral measurements, as will be discussed in Chapter 3, shows that by correcting the frequency shifts in the probe on its co-propagating (return) journey for shifts accumulated during its counter-propagating journey, we can determine the location of the absolute zero timing to within 20% of the laser pulse duration.⁵⁵

CHAPTER 3

INDUCED-PHASE SHIFTS IN COUNTER-PROPAGATING PULSES

In this chapter, I present the first experimental work on time-resolved measurements of induced phase modulation leading to frequency shifts in counter-propagating pump-probe experiments in a glass target with laser pulses of 0.7ps FWHM and peak intensities up to $1 \times 10^{13} \text{W/cm}^2$. Experimental results can only be well explained by including, in addition to the shift induced during co-propagation, a frequency shift proportional to the pump pulse intensity profile during counter-propagation. This additional shift is shown to offer a new approach for mapping of temporal pulse profiles. The effects of pulse asymmetry are also examined. Also, closely related to the studies on the laser-produced plasmas described in the following chapters, these experimental measurements demonstrate a determination of the absolute timing between the pump and probe pulses arriving at the target surface, thus giving additional time information about the plasma formation.

3.1 Introduction

Since the first experimental observations of induced phase modulation effects in bulk glass in 1986² and optical fibers in 1987,⁵⁴ the studies of ultrashort optical pulse propagation in transparent media in the nonlinear regime have stimulated great interest in both theoretical understanding and technological applications of optical materials in 3D imaging,⁵⁶ atmospheric remote sensing,⁵⁷ optical information coding,⁴ pulse compression,⁵⁸ and breakdown studies. For example, when the laser intensity is sufficiently high, information on ionization mechanisms and thus plasma formation or breakdown in solids can be obtained through time-resolved reflectivity and transmissivity measurements.^{27,59} At lower intensities, induced phase modulation (IPM) refers to the frequency shifts experienced by a weak probe pulse propagating through a medium whose refractive index is modulated by an intense pump pulse. While the observations of IPM effects have been demonstrated and reported by many researchers in the past several years with co-propagating laser pulses,^{2,54,60-62} interactions between counter-propagating pulses in nonlinear materials until now have remained unexplored.

In this chapter I report a study of induced phase modulation effects on a weak probe pulse counter-propagating with an intense pump pulse of the same pulselength and wavelength. Combined effects of IPM on co-propagating and counter-propagating probe pulses are measured by recording the shifts in the peak frequency component of the spectrum of the probe as a function of the relative time delay between the two pulses. A large difference in the maximum red and blue frequency shifts has been observed and is well accounted for by incorporating an additional phase change during the counter-propagating journey of the probe in an analytical frequency-chirp model.⁶³ The induced phase in the counter-propagating probe is cumulative in time as the probe pulse passes

through the pump pulse, and it is analogous to pulse walk-off effects described in previous studies.⁵⁴

3.2 Experimental Results

FIG.3.1: Schematic of the pump-probe experiment on IPM effects. The transparent target is also coated with an anti-reflection film to suppress reflection on the rear surface.

A similar detailed schematic of the pump-probe experiment has been described in Chapter 2. Briefly, laser pulses of 0.7ps FWHM at 580nm and about 1mJ per pulse are split into a pump pulse with peak intensity up to $1 \times 10^{13} \text{W/cm}^2$ and a probe pulse with a non-perturbative intensity of $2 \times 10^{10} \text{W/cm}^2$. The pump beam is focused to a $75 \mu\text{m}$ spot at normal incidence onto the front surface of the target. The orthogonally-polarized probe of $25 \mu\text{m}$ focal spot size is incident from the backside at 19° internal angle to the target normal and centered on the pump focal spot (Fig.3.1). This relative beam geometry fixes the interaction length at $L = 120 \mu\text{m}$. By keeping the interaction length L short, the transverse profile of the pump pulse can be assumed constant over L ($\ll$ Rayleigh length

= 7mm), and other nonlinear effects such as stimulated Raman scattering⁶⁴ and temporal broadening due to group velocity dispersion⁴ can all be neglected. The probe pulse reflection, after counter- and co-propagating with the pump pulse in the interaction region, is sent to a grating spectrometer (0.33m, 1200 lines/mm) coupled to a multichannel analyzer (0.6Å per pixel). The glass target is 1mm thick and is coated with an anti-reflection film on its rear surface to suppress probe light reflection from that surface. The target is mounted on a computer-driven translation stage so that on each shot the laser pulses fire on a fresh part of the target while the overlap conditions of the two beams are maintained on each shot.

Spectra of the reflected probe light are displayed in Fig.3.2. Each curve is an average of about 150 shots. The reference spectrum of the probe without the pump pulse (dashed curve) shows a 5Å FWHM sitting on a weaker 13Å wide component centered about the central wavelength. Shot-to-shot spectral jitter is less than 0.3Å. A variable delay line is used in the pump beam to adjust the relative timing of the two pulses for time-resolved measurements. The solid curves are representative red and blue shifted spectra of the probe when it arrives before and after the pump pulse, respectively. In addition to the shift of the peak frequency component, there are also changes in the spectral structure. Figure 3.3 shows the measured shifts (circles) of the peak frequency component in the reflected probe spectra as a function of time delay. The ratio of the maximum red and blue shift is 1.8. The maximum red shift is observed to increase linearly with peak pump intensity (see Fig.3.4), even up to $1 \times 10^{13} \text{W/cm}^2$. At intensities $> 10^{13} \text{W/cm}^2$, an underdense plasma is formed behind the surface which results in appreciable absorption of the probe light,²⁷ (see also Chapter 5), and possible complications from plasma-induced frequency shifts.⁶⁵

FIG.3.2: Spectra of the reflected probe light. Reference spectrum (dotted curve) without intense pump pulse. Solid curves are for early ($\tau_d = -0.1$ ps, right) and late ($\tau_d = +0.7$ ps, left) probe pulses with a peak pump pulse intensity = 4.5×10^{12} W/cm².

FIG.3.3: Measured (circles) and calculated (curves) induced frequency shifts in probe pulses as a function of time delay. Dashed curve is calculation using symmetric Gaussian profile for co-propagating journey only. Dash-dot curve is for counter-propagating only, and solid curve is for both journeys. Dotted curve is calculation for both journeys using an asymmetric pulse profile of Gaussian with 0.24ps risetime and 0.46ps falltime.

FIG.3.4: Measured maximum red shifts (circles) as a function of peak pump intensities. The line is a least squares best fit whose slope determines, as discussed in text, the nonlinear refractive index.

3.3 Theoretical Model & Simulation

The observed large difference in the maximum red and blue induced-frequency shifts is due to the combination of induced phases from the counter- and co-propagating journeys of the probe pulse. In the coordinate τ comoving with the probe pulse, the time dependent refractive index seen by the probe is given by⁶⁶

$$n = n_0 + \gamma I_1(\tau) \quad (3.1)$$

where $\gamma = 8\pi n_2/3cn_0 \approx 2.8 \times 10^{-3} n_2/n_0 (\text{cm}^2/\text{W})$ includes a factor of 2/3 to account for cross interaction between the two orthogonally-polarized pulses.⁶⁷ $I_1(\tau)$ is the instantaneous pump intensity, and $n_0 = 1.52$ and $n_2 = 0.9 \times 10^{-13} \text{ esu}$ ⁶⁸ are the linear and

nonlinear refractive indices, respectively. Considering a δ -like slice of the probe pulse entering the interaction region, the phase change induced on the probe is

$$\Delta\Phi(\tau_d) = a_i \int_{\tau_d - 2n_o L / c}^{\tau_d} I_1(\tau) d\tau + a_r I_1(\tau_d) \quad (3.2a)$$

and the instantaneous wavelength shift is given by

$$\Delta\lambda(\tau_d) = \frac{\lambda}{\omega} [a_i \{I_1(\tau_d) - I_1(\tau_d - 2n_o L / c)\} + a_r \frac{d}{d\tau} I_1(\tau_d)] \quad (3.2b)$$

In these equations, we can ignore the effects of spatial and temporal breakup of the pump pulse as the experimental B-integral is always kept below a value of 3.⁶⁹ The first and second terms describe the induced shifts seen by the probe on counter- and co-propagating paths, respectively. τ_d is the time delay between the two pulses where negative (or positive) τ_d corresponds to early (or late) arrival of the probe relative to the pump, and $\tau_d = 0$ is when the peaks of the two pulses overlap at the surface. The constants $a_r = 2\pi L \gamma / \lambda$ and $a_i = (c/2n_o L) a_r$ represent the phase change per unit pump intensity over the interaction length and phase change per unit pump fluence over the interaction time, respectively. Note that we can ignore pulse walk-off during the return probe journey because of negligibly small normalized group velocity mismatch, i.e. $\Delta v_g / v_g = 1 - \cos 19^\circ \approx 0.05$ over a short interaction length. Similarly for the incoming journey, the factor of 2 in $c/2n_o$ is approximately the normalized group velocity mismatch and accounts for the halved interaction time between the two pulses. The spectral profiles of the reflected probe light are obtained from the Fourier transform or convolution with a probe pulse with a slowly varying envelope, $E_2(\tau)$, and assuming a transform-limited input spectrum ($\delta\nu\delta\tau = 0.36$) :

$$\overline{E}_2(\Delta\omega, \tau_d) = \int_{-\infty}^{+\infty} E_2(\tau) e^{i\Delta\Phi(\tau + \tau_d)} e^{-i\Delta\omega\tau} d\tau \quad (3.3)$$

where $\Delta\omega = \omega - \omega_0$, and ω_0 is the peak frequency component. The shifts in the peak frequency component are then determined by the equality:

$$\frac{d}{d\Delta\omega} |\overline{E}_2(\Delta\omega, \tau_d)|^2 = 0 \quad (3.4)$$

Numerical integration of Eqn.(3.3) is performed for different time delays and pulsewidths, (see Appendix B for computer codes). Results of three calculations using input symmetric Gaussian pump and probe pulse profiles of 0.7ps FWHM are displayed in Fig.3.3. The dashed curve corresponds to the induced wavelength shifts for the co-propagating journey only. The shift is, as expected from earlier analytical models,⁶³ anti-symmetric with respect to zero time delay. At negative delay time, the probe arrives at the surface before the peak of the pump and co-propagates with the leading edge of the pump where dI/dt is positive, thus experiencing a red shift. Conversely, the probe is blue shifted when it arrives after the peak and interacts with the trailing edge of the pump pulse where dI/dt is negative. When the probe co-propagates and tracks the pump precisely at the peak ($\tau_d=0$) it suffers no net frequency shift. These results are similar to the previous experimental results obtained in optical fibers.^{54,57} The dash-dot curve shows the induced shifts accumulated during counter-propagation alone. For early time delay the probe samples more of the leading edge of the pump pulse as the probe and pump pulses cross, resulting in red shifts. At $\tau_d = 0.6$ ps, the peaks of the probe and pump pulses overlap at the midpoint of the interaction region, and hence the probe samples equal amounts of the leading and trailing portions of the pump pulse, leading to no net shift. Finally, at late time delay, the probe predominantly samples the trailing portion of the pump pulse, thus yielding blue shifts. The solid asymmetric curve is the combination of shifts induced

during both journeys, showing good agreement with the observation of shifts at delays $\tau_d < 1\text{ps}$. The discrepancy at later delays may be due to plasma formation during the trailing edge of the pump²⁷ as suggested by a large decrease in the reflected probe signal. The evidence, timing and effects of the plasma formed in the bulk region behind the target will be shown in Chapters 5 and 7. We note that absolute timing between the two pulses can also be obtained with 0.1ps accuracy from the fit to the data shown in Fig.3.3. The effects of pulse asymmetry on the data fitting were also examined by performing the same calculations with various pulse risetimes and falltimes. For example, using an asymmetric Gaussian pulse with risetime of 0.24ps and falltime of 0.46ps the calculation for co-propagation alone still yields a value close to one (1.1) for the ratio of maximum red and blue shifts. The dotted curve in Fig.3.3 is the asymmetric result for both journeys. Such an asymmetric pulse does not significantly affect the ratio of maximum red and blue shifts but does predict a 0.2ps shift in the inferred absolute timing which we take to be another measure of the timing uncertainty. In Fig.3.4, a linear least squares fit to the data points yields a slope of $7.9 \times 10^{13} \text{Åcm}^2/\text{W}$, and after using Eqn.(3.2b), $n_2 = 0.75 \times 10^{-13} \text{esu}$, which is 80% of the tabulated value.

3.4 Summary and Implications

In conclusion, I have reported measurements of induced shifts in reflected probe pulses that counter-propagate and co-propagate with an intense pump pulse. The results show a large difference between the maximum induced red and blue shifts which cannot be explained solely by the previously known analytical model for co-propagating pulses. When the contribution to phase shifts from the counter-propagating journey is included, which corresponds to the case of maximum possible pulse walk-off,^{57,63} the calculations show good agreement with the experimental measurements. We also have shown that induced phase modulation can be used to determine absolute timing between two

counter-propagating pulses within 20% of the pulselength, which is impossible with traditional non-collinear autocorrelators.⁷⁰ Experiments in which transmitted rather than reflected probe pulses are spectrally resolved should isolate frequency shifts from counter-propagating pulses (dash-dot line in Fig.3.3) and are natural extensions of the current work. In addition, experiments with shorter counter-propagating probe pulses to suppress convolution effects, and interaction lengths longer than $c\tau_1/2n_0$ to separate Stokes and anti-Stokes shifts should directly map the pump profile, including any asymmetry.

Both experimental and theoretical results presented in this chapter demonstrate that the induced phase modulation effects associated with the medium refractive index non-linearity (n_2) can be studied at high intensity, up to $1 \times 10^{13} \text{W/cm}^2$. This is done by reducing the interaction length to minimize other nonlinear effects such as stimulated Raman scattering, self focusing or group velocity dispersion reshaping of the laser pulse. Hence, unlike previous work on induced phase modulation where long nonlinear media and much lower intensity have been used, we can single out the IPM effects and therefore simplify in the analysis.

CHAPTER 4

FORMATION OF SUPER-CRITICAL SURFACE PLASMA LAYER BY FEMTOSECOND LASER PULSES

As a transparent target is irradiated with a high intensity laser (pump) pulse, very quickly a high temperature and super-critical or overdense plasma layer is formed at the target surface, making the target highly reflective like a metallic surface. Owing to the heavy masses of ions, plasma hydrodynamic expansion into the surrounding vacuum takes some time to initiate. As the plasma undergoes expansion, its spatial extent grows larger, and the plasma becomes more diluted and transparent to the laser light. Consequently, the surface plasma becomes more absorptive. In this chapter, I present experimental results that confirm the formation of the surface super-critical plasma layer. In addition, the data provide the plasma temperature and the characteristic time scales for the plasma formation and its subsequent expansion into the surrounding vacuum. Calculations using the Saha equations are also shown to determine the relation between the temperature and ionization state of the plasma. Finally, a detailed discussion of the advantages and disadvantages of the frontside probing will be given.

4.1 Time-Resolved Measurements of Frontside Probe Light

FIG.4.1: Simultaneous scans of single shot time-resolved reflectivity and transmissivity measurements by frontside probing with (a) uncoated glass and (b) fused quartz. The pump laser parameters: Peak intensity = $5 \times 10^{14} \text{ W/cm}^2$, FWHM length = 150 fs.

The schematic of the pump and frontside probe experiments has been detailed in Chapter 2. The data were taken with the femtosecond laser. Figure 4.1a shows

simultaneous scans of reflectivity (solid dots) and transmissivity (open dots) as functions of time delay between the strong pump and frontside probe pulse for uncoated glass targets. Each experimental data point is a single shot measurement. The data have been first divided by the incident energy. From the reflection data we then subtracted an average 0.20% of the incident probe light for diffuse pump light leaked into the reflection diode (PD2). The data were then normalized to an absolute value of 4.2% reflectivity for the cold target. Similarly, subtracting an average 4.0% for diffuse pump light leaked into the transmission diode (PD3), transmission data were normalized to a value of 95.8% transmissivity for the cold target. This transmissivity value corresponds to the target which is overcoated with an anti-reflection thin film on the rear surface to avoid reflection from that surface (Fig.1.1). The small fluctuations in the data points are attributed to fluctuations in the pump stray light which do not track the pump energy and to background electronic noise in the boxcar integrators. The zero delay position was inferred from the induced phase modulation experimental results and analysis that were discussed in Chapter 3.

FIG.4.2: Steep gradient of the overdense surface plasma and its scale length increases as the plasma expands

In both data scans, the plateaus during early or large negative delay time correspond to values for the "cold" target where the probe arrives earlier and sees the targets undisturbed by the pump. Following the plateaus, the probe transmission curve drops rapidly and then turns off completely. At the same time, the probe reflection curve rises

with a fast $1/e$ rise time of $\approx 1\text{ps}$ which is longer than the transmission falltime of $\approx 0.2\text{ps}$. The reflection probe then undergoes a much slower decay with a $1/e$ fall time of 6ps . These times are convolved over the finite duration of the probe pulse. After deconvolution, the characteristic time scale for production of the super-critical plasma ($N_{\text{cr}} = 3 \times 10^{21}/\text{cm}^3$) layer is $\tau_{\text{sup}} \approx 0.15\text{ps}$, and $\tau_{\text{exp}} \approx 6\text{ps}$ for hydrodynamic expansion of the plasma. During the transient time, τ_{sup} , a reflecting plasma layer begins to be formed at the surface, and its density increases rapidly to the critical density N_{cr} level and reaches solid state density (10^{23}cm^{-3}). Consequently, transmission of the probe light is decreased quickly and completely stopped, and the probe reflection is strongly enhanced as the surface becomes more metallic-like.^{16,71,72} Since the ions in the material are typically about 4×10^4 times heavier than the electrons and laser energy of the pump pulse is delivered to initially heating of the small mass free electrons, it takes some time for the ions to gain sufficient kinetic energy through collisions with energetic electrons to move or for the plasma expand. Thus, the reflection risetime of 1ps determines the time it takes for the plasma expansion to initiate; and it signifies the ion-electron collisionality or characteristic energy transfer time between ion-electron collisions. As the surface plasma begins to expand after the heating (pump) pulse, the scale length in the underdense region ($< 3 \times 10^{21}/\text{cm}^3$) increases as shown in Fig.4.2.^{10,65} The plasma then becomes more dilute and thus absorptive which results in larger absorption on both arrival and return journeys of the reflected probe light. The reflection falltime of 6ps is thus related to the spatial extent of the plasma expansion; and it is the time where the expansion becomes appreciable. Following the fall, the probe reflection remains roughly at a constant level as the plasma cools off and recombines or vanishingly diluted in the front of the target, leaving the target surface annealed. At much later delay, the reflectivity and transmissivity are found to be similar to that of the undisturbed target, albeit with some small decrease ($\approx -0.8\%$ in reflection and $\approx -5.0\%$ in transmission) due to asperity of the

annealed surface. Figure 4.1b shows the frontside probe data for fused quartz. The data show little or essentially no significant difference with that of glass.

FIG.4.3: Simultaneous scans of single shot time-resolved reflectivity and transmissivity measurements by frontside probing with (a) glass and (b) fused quartz targets overcoated with carbon films of 300Å, showing little or no significant difference from that of uncoated targets. The pump laser parameters: Peak intensity = $5 \times 10^{14} \text{ W/cm}^2$, FWHM length = 150 fs.

Figure 4.3 displays the frontside probe data for (a) glass and (b) fused quartz overcoated with 300Å thick carbon films. The reflectivity and transmissivity levels of undisturbed coated targets are: $R=7.7\%$ and $T=78\%$ for C-coated glass, and $R=7.0\%$ and $T=78.1\%$ for C-coated fused quartz. The data are similar to that of uncoated targets, indicating very little or no noticeable effect of the 300Å thick carbon films.

FIG.4.4: Blue frequency shifts in frontside probe light is proportional to the plasma expansion velocity, taken with fused quartz target overcoated with 300Å carbon film. The pump laser parameters: Peak intensity = $5 \times 10^{14} \text{ W/cm}^2$, FWHM length = 150fsec.

Formation and subsequent expansion of the super-critical surface plasma layer are further confirmed by the time-resolved measurements of frequency shifts in the frontside probe light that interacts with the expanding surface plasma. The blue shifts, shown in Fig.4.4, are proportional to the velocity of the reflecting surface which acts like a reflective mirror moving toward the probe light source. Each data point is a average of 90 laser shots and is the shifts of the center-of-gravity or medium frequency component in the spectra of the puls, i.e. $\Delta\lambda = \lambda_o - \lambda_I$, where λ_I is given from the equality

$\int_{-\infty}^{\lambda} I(\lambda) d\lambda / \int_{-\infty}^{\infty} I(\lambda) d\lambda = 0.5$. Statistical uncertainty in each data point is about $\pm 0.8 \text{ \AA}$, as

indicated by the error bar. The data indicate again that formation of the overdense surface plasma during the heating by the pump pulse takes place rapidly, and its expansion is strongest when the blue shift is maximized, about $7 \text{ \AA}$. After the laser heating, the expansion slows down as the plasma cools off and the expansion velocity is reduced to $\sim 10^6 \text{ cm/sec}$. At this velocity plasma expansion becomes more difficult to detect since the corresponding blue shift is less than $1 \text{ \AA}$, below the experimental uncertainty.

4.2: Discussion and Calculations of Plasma Parameters

The blue shifts in the frontside probe light reflected from the expanding surface plasma are related to the plasma ion acoustic or sound velocity c_s by, using an isothermal plasma expansion⁶⁵ :

$$\Delta\lambda = -2\lambda \cos \theta \frac{c_s}{c} \left\{ \left(\ln \frac{N_e}{N_{TP}} - 1 \right) + 2(1 - \ln 2) \right\} \quad (4.1)$$

In the above, k and c are the wave number and the speed of the laser light, respectively. The wavelength $\lambda = 616 \text{ nm}$. N_0 is the electron density at solid state, and N_{TP} is the density of the plasma at the turning or reflecting point, $N_{TP} = N_0 \cos^2 \theta$. $\theta = 19^\circ$ is the incidence angle of the probe beam. The first term in the bracket corresponds to motion of the reflecting point (Fig.4.2); and the second term accounts for the shift due to the density increase in the underdense region. The factor of 2 in the equation is due to the plasma expansion toward the probe beam. The isothermal model is a good approximation because of the high thermal conductivity in the low density region of the expanding plasma. Using $N_0/N_{TP} \approx 100$ (a typical ratio is often used in previous works^{10,50,73} and

will be justified later), the maximum blue shift $7\text{\AA}$ in Fig.4.4 corresponds to a maximum sound velocity $c_s \approx 5 \times 10^6 \text{cm/s}$. This velocity as will be shown later in Chapter 6 is six times smaller than the supersonic velocity of a ionization front which is supported by electron thermal transport and travels into the transparent bulk region behind the surface plasma.

The maximum velocity of the plasma expansion is given by the isothermal plasma expansion,⁶⁵ i.e. $V_{TP} = c_s (\ln[N_o / N_{TP}] - 1)$, which is about the same as that given by the isotropically adiabatic plasma expansion:^{13,73}

$$V_{TP} = c_s (\ln[N_o / N_{TP}] - 1) \approx 2c_s / (1 - \gamma) \quad (4.2)$$

where γ is the ratio of specific heats of the plasma fluid and is ≈ 1.7 .^{73,74} The maximum expansion velocity is about $1 \times 10^7 \text{cm/sec}$, yielding a scale length of approximately $\tau_{\text{exp}} V_{TP} = 6000\text{\AA}$ for the expanding surface plasma.

The product $Z_{av} T_e$ of plasma temperature and average ionization state is related to the sound velocity through the equality given by Eqn.(E3) in Appendix E. Using $A=20$ as an average atomic number, we find the product $Z_{av} T_e \approx 208$.

The average ionization Z_{av} state of the plasma is a function of the electron temperature and densities of various atomic species. In order to calculate Z_{av} at a given temperature, we use a local thermodynamic equilibrium model (LTE)^{13,75} in which ionization state of each atomic species is in thermal equilibrium with others and is determined by the Saha equation. The use of the LTE model is justified by the fact that at solid density the collisional rate is predominant over the radiative rates, thus making the electron population quickly thermally equilibrated. From the observed risetime of the

reflectivity data and also calculation of equilibration time $\tau_{e-e} \approx 10$ fs using Eqn.(E5), we can safely conclude that the electron population attains Maxwellian distribution in a time $\ll 1$ ps. The Saha equations for the ion distribution of each atomic species is given by:

$$\frac{N_e N_{z+1}}{N_z} = \alpha T_e^{1.5} \text{Exp} \left(-\frac{I(z+1)}{T_e} \right) \quad (4.3a)$$

and

$$N_a = \sum_z N_z \quad ; \quad N_e = \sum_z z N_z \quad ; \quad z = 0, 1, 2, \dots, Z \quad (4.3b)$$

where $\alpha = 6 \times 10^{21} \text{cm}^{-3} \text{eV}^{-1.5}$ is proportional to the ratio of electronic partition functions (g_{z+1}/g_z) which is always of the order of unity and is taken equal to unity. I_z and N_z are the atomic ionization potential energy and number density of z^{th} ions, respectively. N_a and N_e are number densities of neutral atoms and electrons. Equations.(4.3) and (4.4) when applied to multiple atomic materials SiO_2 with $Z_{\text{Si}}=14$ and $Z_{\text{O}}=8$ result in a system of 26 transcendental equations, posing an extremely complicated and time consuming task.

For practical purposes of this chapter, I briefly present now a simple algorithm for calculation of the average ionization state based on the approximate method which has been described in great detail by Raizer.⁷⁶ The calculation is based on two fundamental assumptions: The ionization potential energy is treated as a continuous function of ionization state n , and the Saha equations are replaced by a differential equation $dN_z/dz = N_{z+1} - N_z$ to transform the set of 26 equations into a set of two approximate algebraic equations. Figure 4.5 shows the tabulated ionization energies of oxygen (open circles)

and silicon (closed circles) atoms as functions of ion charge state,⁷⁷ and the best fit (solid) curves. The functional form of the best fit ionization energy curves is

$$I(z) = a_1 \left[\text{Tanh}(a_2(z - a_3)) + \text{Tanh}(a_2 a_3) \right] + a_4 z \quad (4.4)$$

where the fitting parameters a_1, a_2, a_3, a_4 are 414.5eV, 1.45, 6.8, 19.0eV for oxygen, and 1063.8eV, 2.8, 12.57, 37.0eV for silicon, respectively. The average ionization states of oxygen Z_O and silicon Z_{Si} are determined from the reduced set of two equations,¹³ assuming a thermal equilibrium between atomic oxygen and silicon species:

$$Z_{av} = c_O Z_O + c_{Si} Z_{Si} = \frac{\alpha}{N_a} T_e^{1.5} \text{Exp} \left[-I_O(Z_O) / T_e \right] \quad (4.5a)$$

$$Z_{av} = c_O Z_O + c_{Si} Z_{Si} = \frac{\alpha}{N_a} T_e^{1.5} \text{Exp} \left[-I_{Si}(Z_{Si}) / T_e \right] \quad (4.5b)$$

where $c_O = 2/3$ and $c_{Si} = 1/3$ are the relative concentrations of oxygen and silicon atoms, respectively. Figure 4.6 display the results of Z_O, Z_{Si} and the average ionization state Z_{av} of the Si^{+2}_2 plasma as functions of the electron temperature. By locating the point of $Z_{av} T_e = 208\text{eV}$ inferred from the frequency shift measurements above, we have estimates for the plasma temperature and ionization state, namely, $T_e \approx 46\text{eV}$ and $Z_{av} \approx 4.5$. This corresponds to a electron density about 100 times greater than the density at the turning point.

FIG.4.5: Ionization energies as functions of ion charge states. Circles are tabulated values, and solid curves are best fits.

FIG.4.6: Calculated average ionization state of SiO_2 plasma as function of temperature.

4.3 Summary and Limitations

Time-resolved measurements of amplitude (R and T) and phase (frequency shifts) information in the frontside probe light interacting with the surface plasma and its expansion confirm that, upon irradiation of a target with a high intensity laser pulse, a super-critical plasma layer is produced very quickly at the target surface which becomes highly reflective and stops transmission of the laser probe light. The characteristic time constant for formation of the overdense plasma layer is about 0.15ps which is the same as the laser pulse duration as expected. Because of the heavy mass of the ions, the plasma expansion and cooling takes about 1ps to initiate, and $\approx$ 6ps to become significant. The spatial extent of the expanded plasma is about 0.6 μ m. The maximum blue shift in the reflected probe light gives values of $\approx$ 46eV and 4.5 for the plasma temperature and average ionization state, respectively. The experimental results obtained with coated and uncoated glass and fused quartz have been presented to show that effects of the coating films and impurities in the glass targets are not distinguishedly noticeable in the frontside probe signals. This is so because the overdense surface plasma is impenetrable to the frontside probe light and the plasma expansion is predominantly absorptive to the incident and reflected frontside probe light.

Complete analysis of the frontside probe data requires knowledge of the nonlinear mechanisms in which the laser light interacts with the expanding plasma. These include competition between laser pondermotive force and plasma thermal pressure,⁷⁸ stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), density profile steepening and rippling,⁷⁹ and many other nonlinear processes that are associated with long scale length ($L \geq 1000\text{\AA}$) plasma.⁵² This thus presents a rather complicated analysis which lies beyond the purposes of this research work. However, for the conditions of the present experiments, where laser pulsewidth is much longer than the electron-electron collision

time and a Maxwellian distribution for electrons is established, the above calculation using the Saha equations is a good approximation for the plasma temperature and ionization state.

In addition, the frontside probe light, as demonstrated by the data, does not penetrate far beyond the critical density surface, offering no information about the bulk region whose property changes are critical for inference of the energy transport mechanisms. On the issues of laser absorption and hence x-rays emitted from the plasma, because of the overdense surface layer, laser photons that otherwise would continue to be coupled into the plasma and converted into x-ray photons are strongly reflected and prevented from interacting with the plasma. Furthermore, the data presented above clearly show the effects of overdense surface plasma and its slow cooling and expansion into the surrounding vacuum. As the plasma expands, it becomes more diluted and transparent to the laser light. It thus enhances absorption of the probe light in the underdense region and reduces number of laser photons accessible to the plasma region in the bulk region near the critical density. As a result, most of laser-plasma coupling takes place in the underdense region. For these reasons, lengthening of the duration of x-ray burst emitted from the plasma when the pump pulse is long or accompanied with a high intensity prepulse or postpulse has been observed.²⁶

As you see, these are just a few challenges and difficulties in the pump and frontside probe experiments that we now come to realize in the course of searching for the right approach to better understanding of laser-matter coupling and important transport mechanisms in laser-produced plasmas.

CHAPTER 5

FORMATION OF SUB-CRITICAL BULK PLASMA

As was shown in the previous chapter, upon irradiation of a target a super-critical plasma is quickly formed at the target surface, and this plasma keeps the frontside probe light from interacting with the bulk region behind the surface. To provide complementary information about the plasma formation and answers as to how the laser energy is coupled and transported in the bulk transparent solids, time-resolved measurements using a probe light pulse incident from the backside are presented. Backside probing light directly interacts with the bulk region, and at the same time prevents complications due to plasma expansion that was seen with frontside probing. The reason for these advantages is that the plasma, as seen from the backside, is enveloped by a well-defined solid surrounding it and experiences essentially no hydrodynamic expansion. The results taken with both picosecond and femtosecond lasers reveal two competing mechanisms: One is highly absorptive due to a bulk underdense plasma formed behind the target surface, and the other is highly reflective due to an overdense plasma layer at the surface (Chapter 4). A simple self-consistent and analytical model, similar to the avalanche model, is proposed, leading to both time dependent and time-integrated solutions to the evolution of plasma density profiles in the transparent material. Calculations of the probe light interacting with this plasma show that excellent agreement with experimental measurements can only be obtained by including a contribution from the bulk plasma formed behind the surface. Experimental measurements with carbon-and gold-coated targets are also shown to illustrate elimination of the bulk plasma.

5.1 Time-Resolved Measurements of Backside Probe Light & Discussion

Detailed description of the experimental measurements has been given in Chapter 2. Figure 5.1 shows simultaneous scans of reflectivity (a) and transmissivity (b) as functions of relative delay time for uncoated glass targets taken with the picosecond laser. The target was not coated with an anti-reflection thin film on its rear surface. Each experimental data point is a single shot measurement. The data have been first divided by the incident energy. From the reflection data we then subtracted an average 0.15% of the incident probe light for diffuse pump light scattered into the reflection diode (PD3), and 4.0% for the probe light reflected from the undisturbed backside surface. The data were then normalized to an absolute value of 3.7% reflectivity for the cold target. Similarly, subtracting an average 14.0% for diffuse pump light scattered into the transmission diode (PD2), transmission data were normalized to a value of 92.3% transmissivity for the cold target. This transmissivity value corresponds to the target without an anti-reflection film. Absolute zero-time-delay position is determined from the induced phase modulation experiments with the same experimental conditions except that the pump intensity level is attenuated below the plasma formation threshold (see Chapter 3). Note that zero timing is when the peaks of two pulses overlap at the front surface.

In both data scans, the plateaus during early or large negative delay time correspond to values for the undisturbed target where the probe arrives prior to the pump. Following the plateaus, both curves exhibit a transient fall time of about 2ps. The decrease in reflection data occurs about 0.5ps before the decrease in transmission. At late delay time, both reflection and transmission of the probe again show constant levels. The small fluctuations in the data points are attributed to fluctuations in the pump stray light which does not track the pump energy and to background electronic noise in the boxcar integrators. At much later delay, many seconds after plasma formation when the plasma

has completely recombined leaving the surface annealed, the reflectivity and transmissivity are found to be similar to that of the undisturbed target, albeit with some small decrease ($\approx -0.9\%$ in reflection and $\approx -7.0\%$ in transmission) due to asperity of the annealed surface.

FIG.5.1: Simultaneous scans of (a) transmission & (b) reflection taken with uncoated glass (open dots). Solid curves are calculations discussed in section 5.2. Arrows mark the beginnings and endings of the transients corresponding to points of 10% difference from the plateau levels. Dashed curves are that of surface plasma without absorptive effect by the bulk plasma. Laser parameters: Peak intensity: $2 \times 10^{13} \text{W/cm}^2$, Pulselength: 1psec.

The temporal behavior observed in the probe reflection and transmission can be qualitatively understood by considering the sequence of events as the pump pulse arrives at the target surface:

At early time, or during the leading edge of the pump pulse where the intensity at the surface is low, nearly 96% of the light is transmitted through the front surface. As the transmitted pump light propagates into the bulk region, it is absorbed and ionizes the transparent material, resulting in a bulk underdense plasma behind the surface. Since the density of the undercritical plasma ($< N_{cr} = 3.3 \times 10^{21} \text{cm}^{-3}$) is much below solid density ($N_a = 6.6 \times 10^{22} \text{cm}^{-3}$), ionization of neutral atoms is the main contribution to the formation of the plasma.^{13,80} The velocity of the ionization front should be $\approx cn_0/n_g^2$, where c is the speed of light, and the refractive index in the region swept through by the ionization front is $n_0 \approx \sqrt{(n_g^2 - N_e/N_{cr})} \approx n_g = 1.52$. The spatial steepness of the leading edge of the ionization front depends on the risetime of the laser profile at the intensity level which produces such an electron density. At some time t_{cr} , there is a critical density plasma layer formed at the surface. As will be shown in the next section, the critical time t_{cr} and the spatial extent of the plasma at t_{cr} will be quantified using an avalanche model. When this plasma layer becomes thick enough ($\approx 0.1 \mu\text{m}$), it completely stops further pump pulse transmission as was evidenced from frontside probe data shown in Chapter 4. As a result, the remaining pump light is kept from further interacting with the bulk region behind the surface plasma layer, and instead, deposits its energy at the surface plasma layer, quickly producing a thin region of highly reflecting, solid density plasma.¹⁶ The transmitted portion of the pump pulse continues deeper into the target, ionizing until it exits at the rear surface. Thus, at late time there are two competing mechanisms for the backside probe pulse: One is highly reflective due to the formation of a steep overdense

layer at the surface, the other is strongly absorptive due to a bulk underdense plasma produced behind the surface.

In view of this picture, we explain the temporal behavior of the probe data as follows. At early delay, prior to the critical time t_{cr} , there is a small underdense plasma which causes little absorption in the transmitted probe during its incoming journey to the front surface. The probe light reflected from the surface, however, will co-propagate with the ionization front and hence suffer additional absorption over a maximum interaction length of $100\mu\text{m}$ on its return journey. This accounts for the earlier decrease in the reflected light. Thus, the time at which the reflection transient begins should determine, after convolution with the probe pulse profile, the time at which a plasma of an appreciable density is formed by the leading edge of the ionization front. As will be seen later, this density of about 10^{18}cm^{-3} reduces probe reflection by 10% from the value of the cold target. At the critical time t_{cr} , the probe transmission is completely turned off by formation of a critical density plasma and subsequently a surface overdense layer. By contrast, the probe reflection may be enhanced by the surface plasma layer, only to be heavily absorbed again by the bulk plasma on its return journey. At the start of the reflection transient, it is the trailing edge of the probe pulse that is absorbed appreciably. The end of the transmission transient occurs when the leading edge of the probe pulse sees the surface critical plasma. Therefore, the time lag of $\approx 2.5\text{ps}$ between beginning of reflection transient and ending of the transmission transient is the estimate of the time lapse, convolved by the probe pulselength, between the formation at the surface of an appreciable density plasma and critical plasma, and this time lapse, as will be shown later, is about 0.14ps . Since the time scales for cooling and hence recombination in the underdense plasma by electron-atom and electron-ion collisions are $>100\text{ps}$,^{13,80,81} these processes can be neglected on the few picosecond time scale of Fig.5.1. Also, measurements with Au-coated targets described below (Fig.5.2) confirm that surface

plasma reflectivity remains approximately unchanged over the time scale of few picoseconds. For these reasons, the final reflection and transmission levels of the probe light should also remain approximately constant as is observed.

FIG.5.2: Simultaneous scans of reflectivity (closed dots) and transmissivity (open dots) for Au-coated glass targets. Unlike uncoated targets, reflection enhancement is seen as an indication of elimination of absorption by an underdense plasma. Laser parameters: Peak intensity: $2 \times 10^{13} \text{ W/cm}^2$, Pulselength: 1psec.

Specifically, to isolate the effects of such a highly reflecting surface overdense plasma, we have performed the same experiments with the glass targets overcoated with an absorptive gold film of $300 \text{ \AA}$ thickness at the front surface. The target was also anti-reflection coated on its rear surface to enhance the contrast in the reflection data between early and late time delay. The use of the Au-coating is to promote larger absorption of the intense pump pulse at the surface, leading to an earlier formation of a surface overdense plasma, hence preventing formation of the bulk underdense plasma behind the coating. An estimate of the critical intensity level at which the Au region reaches critical density gives a value of $\approx 4 \times 10^{11} \text{ W/cm}^2$. As will be seen later, the bulk plasma produced in the

glass by the transmitted pump pulse at this intensity level is of negligible density. The absorption of the cold Au layer is $\approx 12\%$, inferred from values of 64% reflectivity and 24% transmissivity that are measured by the probe. The data, as displayed in Fig.5.2, taken in the same manner described above were normalized to reflectivity and transmissivity of the cold target. The data show a transmission transient time of 2.0ps, about the same as that of uncoated targets. At the same time delay, however, there is a reflection enhancement of nearly 10% of about 2.0ps duration. Following the reflection enhancement is a long reflection decrease with 1/e-decay time of about 8.0ps. This behavior proves that with Au-coated targets we have succeeded in isolating the highly reflective surface overdense plasma layer from large absorption by a bulk underdense plasma. Further detailed discussion of measurements and calculations for coated targets, taking into account for the effects of thermal heat wave^{13,28} to explain the slow decay in reflectivity, will be given in Chapter 6.

FIG.5.3: Simultaneous scans of (circles) transmission & (rhombus) reflection of backside probe light for uncoated glass targets taken with femtosecond pulses. Solid curves are calculations from the model discussed in section 5.2. Arrows mark the beginnings and endings of the transients corresponding to points of 10% difference from the plateau levels. Laser parameters: Peak intensity: $5 \times 10^{14} \text{ W/cm}^2$, Pulselength: 200fsec.

FIG.5.4: Simultaneous scans of transmission (open dots) & reflection (closed dots) of backside probe light for carbon-coated glass targets taken with femtosecond pulses. Reflection increase, due to the thermal effect discussed in Chapter 6, illustrates the disappearance of sub-critical bulk plasma. Laser parameters: Peak intensity: $5 \times 10^{14} \text{W/cm}^2$, Pulselength: 200fsec.

Figure 5.3 shows the data taken in the same procedures described as above with the femtosecond laser. Again, the evidence of the bulk plasma is strongly confirmed by large absorption in the backside probe reflection signals and the early fall in the reflectivity, about 0.2psec prior to that of the transmissivity. Figure 5.4 displays the data for carbon-coated glass target taken with femtosecond pulses. The absorptivity of cold C-coated targets is $\approx 14\%$, inferred from values of 7.8% reflectivity and 78% transmissivity. Reflectivity enhancement reaches a maximum of 22%. The 1/e risetime in the reflection enhancement is about 0.4ps, equal to that of the decrease in the transmissivity data. This time, after deconvolving with the finite duration of the probe pulse, is equal to the pump pulse duration. The slow decay following the maximum reflectivity has 1/e falltime of $\approx 1.5\text{ps}$, which is about 7 times longer than the risetime, about the same ratio that was seen with the picosecond data in Fig.5.2.

5.2 Modified Avalanche Model for Evolution of Bulk Plasmas In Transparent Materials: Simulation Results and Calculation

Early during the rising edge of the strong pump pulse, prior to the critical level where a critical density plasma is formed at the surface, most (or 96%) of the high intensity laser light is transported into the transparent material. Under the influence of this laser field, neutral atoms are ionized. It then gives rise to an underdense plasma, below the critical density, inside the transparent region as the ionization front, created by the high field ionization, travels inward into the target. Since the underdense plasma density is much below solid density, the main contribution to forming the underdense plasma is the ionization of neutral atoms by inverse Bremsstrahlung absorption of the laser photons in electron-atom collisions.^{13,80} Thus, the physical problem involves the transport of the laser photons flux in a lossy medium which is in turn determined by the plasma absorptivity. The problem also involves evolution of the plasma both in time and space as the traveling laser photons are absorbed in the electron-atom collisions. It should be noted that apart from random motions no particle undergoes any directed motion ($u=0$). Also, while the number density of atoms and ions in the target region is conserved, the free electrons are born locally as laser photons are propagating and absorbed. As will be seen in a steady state (constant temperature), no current of free electrons on the direction of the electron density gradient because an electric field would be generated rapidly. For these reasons the commonly used continuity equation in the fluid dynamics is not relevant and can be ignored. Thus, in order to model self consistently the time evolution of the bulk underdense plasma in space and in the short time scale of the experimental measurements as described above, we propose the following equations (in cgs units) :

$$\frac{1}{V_g} \frac{\partial I(x,t)}{\partial t} + \frac{\partial I(x,t)}{\partial x} = N_a \sigma_v(x,t) I(x,t) \equiv \kappa(x,t) I(x,t) \quad (5.1)$$

$$\begin{aligned} (E_i + \frac{3}{2} kT_e) \frac{\partial N_e(x,t)}{\partial t} + \frac{3}{2} kN_e \frac{\partial T_e(x,t)}{\partial t} = N_a \sigma_v(x,t) I(x,t) + \\ \left[-D \nabla^2 (N_e \frac{3}{2} kT_e) + \frac{3}{2} kN_e (T_e - T_a) v_{e-a} \right] \end{aligned} \quad (5.2)$$

Equation (5.1) is the transport equation for a wave packet of laser photons traveling in a transparent medium with a group velocity $V_g \approx c/n_g$ and an absorption coefficient κ equal to the product $N_a \sigma_v$. N_a is number density of neutral atoms. σ_v is the cross section for absorption of a laser photon during an electron-atom collision and is linearly proportional to electron density,⁸⁰ $\sigma_v = bN_e/N_a$, where b is defined as the cross section at solid density. Equation (5.1) describes characteristics of laser pulse propagation and absorption in the transparent medium: As it propagates it is absorbed and attenuated, imparting energy to heating of the free electrons. Consequently, deformation of the pulse profile occurs. Early during the leading edge of the pulse, when the electron density is still low, the effect on the profile is small. At later time, the increasing plasma density has a greater effect on the profile. More details on the characteristics of the laser pulse propagation are given in Chapter 7.

Similar to the initiation of breakdown in solid materials by an avalanche process,^{14,82-84} creation of electron density begins with a small number N_{e0} of free electrons that are generated by multiphoton processes or by readily-ionized impurity atoms. When these initial electrons gain sufficient kinetic energy $3kT_e/2$ from the laser field, they collide with nearby atoms to produce a cascade of secondary electrons. Equation (5.2), similar to that of Harrach's model,⁸⁵ describes evolution of an electron plasma with E_i as ionization energy except for an additional term due to spatial diffusion

of electrons out of the interaction region. On the few picosecond time scale, energy losses due to ambipolar diffusion ($\approx 100\text{ps}$ for a diffusion scale of $1000\text{\AA}$), lateral electron thermal conduction or free streaming diffusion ($\approx 100\text{ps}$),⁸⁰ and the radiative cooling time ($\approx 100\text{ns}$)⁸¹ can be all safely ignored. The second term in the bracket for energy exchange in direct electron-atom collisions can also be neglected by virtue of the long characteristic time ($\approx 1\text{ns}$) for appreciable energy transfer in electron-atom collisions.

According to Harrach's model, the plasma formation and heating process reveal two distinct stages.^{85,86} The first stage is the heating of initially free electrons to some high characteristic temperature, followed by a stage of ionization growth in which collisions between hot electrons and atoms prevent further temperature change. His model gives an estimate of $\approx 10\text{fs}$ for the characteristic heating time and the characteristic temperature of $\approx 1.0\text{eV}$ at 10^{13}W/cm^2 intensity. For our experimental conditions, we therefore neglect the term in $\partial T_e/\partial t$, and assume a maximum final temperature of 1.0eV that is given by the Saha equation (Chapter 4) for $E_i = 9\text{eV}$, $N_e \leq N_{\text{Cr}}$ and $N_{\text{Cr}} = N_a/20$. Another way to justify omission of this term is by examining equation (5.2) without the terms in the bracket. The electron density grows exponentially with a growth rate proportional to the intensity whereas temperature only increases as some power of intensity, and this power dependence is less than one since ionization competes with the heating process. A physical implication of this approximation is analogous to that given by Newstein.⁸⁶ Under the influence of the laser field, at some characteristic time the free electrons gain as much energy from the field as they lose to many cold electrons that are bound to the nearby atoms. Thus, there is a steady state in which further heating is balanced by the rate of energy lost to ionizing neutral atoms. As a result, energy absorbed during the subsequently arriving pump pulse does not give rise to any further significant temperature increase, but instead goes toward the predominant ionization of atoms. For these reasons

we can also then neglect the temperature dependence of the electron-atom collision cross section.

Equation (5.2) preserves the exponential nature of the avalanche model where the ionization rate coefficient is linearly proportional to the laser intensity and electron density is a steeply rising function of intensity. Therefore, the critical threshold $I_{cr} = I(0, t_{cr})$ and critical time t_{cr} are weakly dependent or essentially independent of the initial condition $N_{e0}(x)$. Hence, early during the leading edge of the laser pulse, details of any ionization process other than collisional ionization such as multi-photon, photo-ionization of impurities, F-center formation are unimportant and just serve to produce a number of "priming" electrons $N_{e0}(x)$, initiating the subsequent cascade processes in the interaction region.^{14,82,84}

Interestingly, equation (5.2) with the terms discussed above neglected together with equation (1) are analogous to those of Frantz and Nodvik⁸⁷ for laser amplification except we describe here propagation of a high intensity laser pulse in an absorptive medium, with an absorption that depends approximately exponentially on the laser intensity. Solving the two equations by the method of characteristics with an input laser pulse $I_0(t) = I(0, t)$ at the surface and an initial electron density $N_{e0}(x) = N_e(x, -\infty)$, time dependent solutions to the electron density evolution and laser pulse propagation are, with $I_0(t \geq t_{cr})$ set to 0 to simulate formation of the surface critical plasma, given by (see Appendix A for further algebraic details) :

$$I(x, t) = \frac{I_0(t - x/V_g)}{1 + \text{Exp}(a \int_{-\infty}^{t-x/V_g} I_0(t') dt') \left[\text{Exp}(b \int_0^x N_{e0}(x') dx') - 1 \right]} \quad (5.1a)$$

$$N_e(x, t) = \frac{N_{e0}(x)}{1 + \text{Exp}(-b \int_0^x N_{e0}(x') dx') \left[\text{Exp}(-a \int_{-\infty}^{t-x/V_g} I_0(t') dt') - 1 \right]} \quad (5.2a)$$

where the proportionality constant $a = b/(E_i + \frac{3}{2}kT_e)$.

FIG.5.5: Evolution of electron density behind the transparent target surface (a) 1.2ps laser pulse, (b) with 200fs pulse. Negative (positive) time is when the leading (trailing) edge of the pump pulse arrives at the surface ($x=0$). Zero time corresponds to the peak of the pulse at the surface.

The time evolution, from -3.0ps to +3.0ps, of the electron density normalized to the critical density is shown in Fig.5.5a. The pump pulse (not shown in the figure) is incident onto the target surface ($x = 0$) from the left. Note that zero time refers to the time when the peak of the pulse arrives at the surface. Parameters used for the calculation are: Peak intensity $I_{\text{peak}} = 2.0 \times 10^{13} \text{ W/cm}^2$ with a Sech² profile and FWHM=1.2ps; $T_e = 1.0 \text{ eV}$; $b = 1.2 \times 10^{-17} \text{ cm}^2$. Ionization energy is taken as the energy gap (=9eV) between the valence and conduction bands of the insulator material. We take $N_{e0} = 10^8 \text{ cm}^{-3}$, a typical value often used in previous work on laser-induced damage.^{14,88} This number corresponds to ≈ 10 initial electrons in the interaction volume of $\approx 10^{-7} \text{ cm}^{-3}$.

The critical plasma layer is formed at the surface during the leading edge of the pulse, at $t_{\text{cr}} = -0.46 \text{ ps}$ and $I_{\text{cr}} = 1.3 \times 10^{13} \text{ W/cm}^2$. The calculation shows the time lapse for the density growth at the surface from a density of 10^{18} cm^{-3} to N_{cr} is $\Delta t = 0.14 \text{ ps}$. Prior to the critical time, t_{cr} , the plasma buildup rate is given by equation (5.2), $N_e'/N_e = aI(x,t)$, an increasing function of time up to a maximum at t_{cr} which sets an upper limit for the peak intensity of the transmitted pump pulse. At time $t > t_{\text{cr}}$ an underdense plasma plateau of $\approx 10^{19} \text{ cm}^{-3}$ begins to develop inside the bulk region as the transmitted pump continues to the rear surface ($x = 1000 \mu\text{m}$). The average plateau density is about 330 times below the critical density and about 6000 times less than the solid density.

Figure 5.5b shows similar calculation for electron density evolution with 200fs FWHM Sech² pulses. Again there is a large underdense plasma in the bulk region behind the surface which give rise to large absorption and early fall in the reflection of the probe light as the shown in Fig.5.3. The surface critical density occurs at -71fs during the leading edge of the pump pulse.

FIG.5.6: Ionization rates as a function of critical or breakdown threshold field. Experimental points (open dots) and semiempirical calculation (dashed curve) are taken from N. Bloembergen, 1974,⁸³ for comparison with the spatially averaged rates (solid line) given by equation (5.2). The solid dot corresponds to calculation based on the current experimental conditions. (See Bloembergen, 1974, for a more detailed discussion of the semiempirical curve.)

Comparisons of ionization rates with previous measurements obtained from the laser-induced damage studies on NaCl crystals by laser pulses of 20ps and longer duration are plotted in Fig.5.6 against the critical or threshold electric field.^{82,83} The experimental data (open dots) together with the semiempirical calculations (dashed curve) are taken from Bloembergen, 1974.⁸³ The curve was derived with an assumption that the free electron mobility is independent of the electric field, and it shows the ionization process is neither strictly intensity dependent nor strictly energy fluence dependent. The solid line, resulted from averaging $\alpha I(x, t_{cr})$ over the interaction length for $I_{peak} =$

$2.0 \times 10^{13} \text{W/cm}^2$ and various pulselengths, is energy fluence dependent only and is asymptotically a limiting case to the semiempirical curve in the region of short pulse duration. This is because under the influence of laser pulses of duration comparable with the time between electron-atom collisions, the electron drift velocity is strictly linearly proportional to the laser field. Our maximum ionization rate at t_{cr} (solid dot) corresponding to the current experimental conditions is $4.3 \times 10^{13}/\text{sec}$.

FIG.5.7: Shown in greater detail, electron density profiles and cumulative optical depths as functions of position at times t_{cr} (solid curves) and at $t_{\text{cr}} + 0.5 \text{ps}$ (dashed curves).

In Fig.5.7, we display in greater detail snapshots of electron density at times t_{cr} (solid curve) and $t_{\text{cr}} + 0.5 \text{ps}$ (dashed curve) in the bulk region ($x > 0.1 \mu\text{m}$). The surface overdense layer region (not shown in the figure), not treated by the model after it is formed, is thought to be of order $0.1 \mu\text{m}$, which is many optical skin depths. At t_{cr} the thickness of the bulk plasma region ($\geq 10^{18} \text{cm}^{-3}$) at the surface is $= \Delta t v_g = 28 \mu\text{m}$. The shoulders in the density curves correspond to the moving ionization front whose time dependent slope is sensitive to the pulse risetime near the critical threshold. Also shown

are curves for the cumulative optical depth (defined as $\int^x \kappa(x') dx'$), describing the spatial distribution of the bulk absorption.

FIG.5.8: Profile of the input pump pulse at surface (solid curve) taken as Sech^2 with 1.2ps FWHM, and profile of the transmitted pulse at the rear surface (dashed curve) resulting in shorter FWHM and lower peak intensity. Note the zero point in transmitted pump profile occurs about 0.05ps prior to the critical time $t_{cr} = -0.46\text{ps}$.

The profile of the calculated transmitted pulse (dashed curve) exiting at the rear surface plotted in Fig.5.8 is compared to that of the input pulse (solid curve) at the front surface, showing clearly the timing of maximum absorption of the transmitted pump energy. Note the zero-intensity point in the transmitted pump pulse occurs about 0.05ps prior to t_{cr} . The sudden decrease in the transmitted pump pulse profile due to the rapid buildup of plasma density has been observed previously and used to determine breakdown threshold electric fields with pulses of many picoseconds.^{83,84} At t_{cr} decoupling of the transmitted and reflected pump pulses should be completed by the surface overdense plasma, and subsequently, the pump reflection should be increased strongly which forms the basis of the plasma mirror effects.²⁶ We note that for Au-coated targets the intensity for critical plasma formation ($I_{cr} \approx 4 \times 10^{11} \text{W/cm}^2$) occurs about 1ps

earlier and at an intensity about 30 times lower than that in uncoated targets. Hence, bulk absorption at this level, as is shown in Fig.5.8, is indeed negligible.

We now model the probe pulse absorption. Absorption of the probe light is by e-atom inverse Bremsstrahlung and obeys Beer's law. Considering a δ -like slice of the probe pulse entering the interaction region and reaching the front surface at time t prior to the critical time, $t < t_{cr}$, the probe time-dependent transmission and reflection are given by:

$$T(t) = T_g \text{Exp} \left[- \int_0^{L_i} \kappa(x, t - x / V_g) dx \right] \quad (5.3a)$$

$$R(t) = R_g \text{Exp} \left[- \int_0^{L_i} \kappa(x, t - x / V_g) dx - \int_0^{L_i} \kappa(x, t + x / V_g) dx \right] \quad (5.3b)$$

where T_g and R_g are the transmissivity and reflectivity of the cold glass target, and $L_i = 100\mu\text{m}$ is the interaction length. The second exponential term in R accounts for additional absorption by the reflected probe pulse during its return journey.

For time $t \geq t_{cr}$ transmission of the probe is stopped, and probe reflection is modified by the formation of the surface plasma layer :

$$T(t) \approx 0 \quad (5.4a)$$

$$R(t) = R_s \text{Exp} \left[- \int_0^{L_i} \kappa(x, t - x / V_g) dx - \int_0^{L_i} \kappa(x, t + x / V_g) dx \right] \quad (5.4b)$$

where R_s is the reflectivity of the surface overdense plasma layer. Previous experimental measurements of reflectivity with subpicosecond pulses have shown that surface reflectivity range from 30% to 85% is possible.^{9,17} For example, results from frontside probe data shown in Chapter 4 determine the observed surface reflectivity of about 30%.

Our estimates from Fresnel calculations of the surface reflectivity give a value of $\sim 40\%$ for R_s , which will be treated as a constant in a time scale of few picoseconds as was seen with Au-coated targets in Fig.5.4. The above expressions are for instantaneous values which are then convolved over the finite duration of the probe pulse.

FIG.5.9: Correlated values of surface overdense plasma reflectivity and solid density cross section (open-square curve) that give best fits to the experimental reflectivity & transmissivity data. Also plotted are the corresponding absorption coefficient at t_{cr} (open-circle curve) and at $t \geq t_{cr} + 0.5\text{ps}$ (closed-circle curve).

To obtain least square best fits to the data for uncoated targets shown as solid curves in Fig.5.1, we have used a pulse length of 1.2ps FWHM, 0.2ps longer than the recorded autocorrelation shown in Fig.2.3. The most probable justification for this discrepancy is ascribed to variations in the pump pulse intensity across the focal diameter which translates into a range of critical times across the diameter of the plasma (see Chapter 7). This spatial dependence of intensity was not accounted for by the model. The competition between the bulk plasma and the surface plasma is characterized by a relation between values of b and R_s that produce best fits to the experimental data. In Fig.5.9 calculated values of b are plotted as a function of R_s (open-square curve) which give us estimates

for the lower and upper limit of the cross section b for photon absorption. We note that values of b range within a factor of 3 for a reasonable range in R_s (10-70%). The average value of b of $\approx 1.0 \times 10^{17} \text{cm}^2$ is about five times lower than that predicted by Browne's formula where it was derived for plasma formation in gas.⁸⁰ We attribute this reduction to the band structure and quantum transport effects in SiO_2 solid.⁸⁹ Also, the absorption coefficient of the bulk plasma, averaged over the interaction length, are plotted on the right scale against the values of b at time t_{cr} (open-dot curve) and at time $t \geq t_{\text{cr}} + L_{\text{inO}}/c = t_{\text{cr}} + 0.5 \text{ps}$, the time when the density plateau begins to extend beyond the interaction region (close-dot curve).

Another demonstration of the absorptive effect of the bulk underdense plasma on the measured reflection and transmission of the probe light can be shown by repeating the calculations (dotted curves in Fig.5.1) without absorption by the bulk plasma. As one would expect from this calculation, there is no time lag between the transients of the reflectivity and transmissivity, and more importantly, there is a reflectivity enhancement rather than a decrease as was observed. Thus, had the bulk plasma not been produced in uncoated targets, we would have seen a large reflection enhancement of backside probe light as was observed with Au-coated targets shown in Fig.5.4, C-coated target in Fig.5.5, or a reflection increase similar to those of frontside probe data in Chapter 4.

5.3 Summary and Significance

We have reported time-resolved measurements of the reflectivity and transmissivity of plasmas produced by both intense picosecond and femtosecond laser pulse irradiation of initially transparent targets. The results demonstrate that before the peak of the pulse, there is a large underdense plasma formed in the bulk glass region which gives rise to absorption of the probe light. At subsequent times there is also an overdense plasma layer

formed at the surface which prevents further transmission of the laser light and thus any subsequent increase in the plasma density and temperature in the bulk region. The data show competition of two opposing mechanisms, one is reflection by the surface overdense plasma and the other is absorption by the bulk underdense plasma. Experimental results for targets overcoated with a thin gold film showed that the bulk plasma can be eliminated as predicted. Moreover, related to the unsolved issues of previous works on laser-induced breakdown of optical materials,⁹⁰ our experimental findings of the underdense plasma imbedded in the solid may very well be the direct evidence for avalanche buildup of charge in the bulk region. Time-resolved studies of the surface plasma and the bulk plasma provide dynamical pictures for different processes that lead to surface and bulk damages that have been seen in wide variety of optical materials.^{14,90}

We also have presented a model for plasma evolution in transparent materials with laser pulses of arbitrary shape. The model omits several physical processes, as were justified by their characteristic times that are irrelevant to the time scale of the experiment. These omissions are necessary to render simplicity and analyticity of the model, and at the same time, to retain the physical features similar to some previous models and preserve the characteristics of the well-known avalanche model. Moreover, unlike others, our model supplementally and self-consistently describes the characteristics of the high intensity laser pulse propagation in the target medium as it is absorbed by e-atom inverse Bremsstrahlung, and it determines the growth rate, size and absorptivity of the bulk plasma produced by the laser pulse. Also, it estimates the maximum amount of laser energy that can be transmitted through a transparent medium which is critical in studies of optical breakdown and the technology of high-power propagation in optical materials. The simplicity of the model allows one to perform rapid simulations on personal computers, and open up ways for both time-resolved and integrated predictions

of plasma dynamics in transparent solid materials. In particular, the model is useful for predicting the formation of relativistic underdense ionization fronts which are of current interest for frequency upshifting.^{91,92}

Calculations of the reflected pump energy and pulse profile can also be obtained by energy conservation, but they will be somewhat overestimated since the model does not include absorption of the pump energy by the overdense plasma layer at the front surface and its subsequent expansion into vacuum. These calculations, nevertheless, prove the possibilities for further investigation of time-resolved breakdown or plasma formation in transparent materials. All of these calculated parameters for the pump pulse, except t_{cr} , can be directly obtained from experiments. Information about the critical time, however, can be determined from comparison of the transmitted and reflected pulse profile with the incident pump, for example, by high order autocorrelators. Concerning plasma mirror effects, the above calculations show that recording of the energy and profile of the transmitted pump pulse rather than reflected pump pulse is more meaningful in that they are devoid of complications due to the effects of hydrodynamic expansion at the front surface. Furthermore, since most of the laser absorption is predicted to occur, for examples, in case of picosecond pulses, 0.14ps prior to the critical time, the transmitted pump profile should allow us to determine the critical time and intensity threshold with an accuracy better than half of the picosecond laser pulse FWHM.

On the issues of laser-matter coupling and transport mechanisms, the time-resolved measurements of backside probe light with uncoated glass targets confirms, in addition to the highly reflective overdense plasma layer at the surface, production of a sub-critical bulk plasma. This plasma is so strongly absorptive that the reflected backside probe light is too weak for any additional useful information about the surface plasma and transport mechanisms. As will be seen in the next chapter, the effects of the bulk plasma can also

be greatly reduced by using uncoated targets of pure fused quartz. By utilizing the highly absorptive carbon thin films, the presence of a bulk underdense plasma and effects of induced phase modulation as was discussed in Chapter 3 can be altogether eliminated. These, together with the advantages of backside probing, make it possible and easier to isolate and analyze the effects of electron thermal conduction, as will be shown in great details in the next chapter.

CHAPTER 6

TIME-RESOLVED ELECTRON THERMAL CONDUCTION

In this chapter, I report the most important experimental findings of this research work about energy transport mechanisms in femtosecond-laser-pulse-produced plasmas. In the first section of this chapter, I summarize briefly, from the work presented in the previous chapters, the experimental difficulties in probing electron thermal conduction. The second section shows the experimental results where I have succeeded to isolate the electron thermal conduction mechanism with targets overcoated with a highly absorptive amorphous carbon thin film. These data are clear evidence for a supersonic thermal wave, supported by electron thermal conduction, during the first picosecond after the femtosecond laser irradiation of the target. Formation of a super-critical surface plasma with a rather steep gradient is initially strongly regulated by the dominant electron thermal transport until, at about 1ps later, when plasma expansion begins to set in, a rarefaction wave resulting from plasma expansion takes part in the energy transfer process, competing with electron thermal transport. In section 3, I present a nonlinear heat wave model and an approximate analytical solution to a nonlinear heat diffusion equation to describe evolution of electron plasma temperature and density in both time and space. Analysis and calculations using the model shows (section 4) good agreement with the experimental observations. Finally, I close the chapter with some remarks on the importance of the findings and implications of the calculation.

6.1 Experimental Difficulties

We have learned that (Chapter 3) when the pump laser intensity is below the plasma formation threshold ($<10^{13}\text{W/cm}^2$), the probe light sees an induced phase modulation associated with the nonlinearity in the refractive index n_2 of a glass medium.⁵⁵ This nonlinearity is determined by the electronic nonlinear activity of bound electrons.¹⁴ At higher intensity, neutral atoms are ionized, and the bound electrons become free, giving rise to an induced phase modulation that is a consequence of a negative change in the refractive index through formation of a plasma (see also Chapter 7). As was discussed in Chapter 4, when the plasma formation at the target surface was monitored by frontside probing of an uncoated transparent glass target, the frontside probe yields temperature and characteristic time constants for the surface plasma and its subsequent expansion into the surrounding vacuum. The frontside probe, however, provides no useful information about plasma formation mechanisms and important energy transport. The intrinsic difficulty of this was the overdense surface plasma layer which strongly reflects and prevents the frontside probe light from interacting with the plasma behind the target surface. This problem was then tackled by using a backside probe beam (Chapter 5). The goal was primarily to avoid many complications due to plasma expansion at the front side of the target and directly sense the bulk region behind the surface whose changes in the material properties are necessary for inference of the energy transport mechanisms. This then led us to the findings and confirmation of a large underdense plasma in the bulk target. Another difficulty of probing electron thermal conduction that arose was that the backside probe light signal, after interacting with such a large and strongly absorptive plasma, was too weak for any meaningful interpretation of the important mechanisms.

In order to circumvent the aforesaid difficulties and complications for isolating electron thermal conduction and simplifying the analysis, a transparent pure fused quartz

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target overcoated with a highly absorptive carbon thin film of $300\text{\AA}$ thickness was used. As was demonstrated in Chapter 5 and more details in Chapter 7, the absorptive coating effectively lowers the plasma formation threshold, thus reducing the intensity level and total energy of the transmitted portion of the strong pump pulse. The positive results are that the complex nonlinear effects of the induced phase modulation and formation of an underdense bulk plasma are altogether suppressed. This, together with the advantage (discussed in Chapter 5) of the backside probing, allows the acquisition of experimental results uncomplicated by plasma expansion, hence isolating the effects of thermal transport. It should be noted because of the short time scale during which the experimental data are taken, complications from plasma hydrodynamics or development of a shock wave resulting from plasma expansion can be further simplified. Another important advantage of the absorptive film will be realized more clearly later with the experimental data as presented in the next section.

6.2 Experimental Observations of Electron Thermal Conduction

As is well known so far and was discussed in Chapters 4 and 5, on irradiation of a transparent solid target overcoated with an absorptive thin film, a high temperature and solid density plasma layer is quickly formed at the target surface during the rising edge of the laser pulse.²⁷ As a result, the remainder of the laser pulse is shielded from the bulk region behind the critical density surface ($\approx 3 \times 10^{21}/\text{cm}^3$) where the plasma frequency is greater than the laser frequency. The remainder of the pulse however continues to interact with the surface plasma and deposit its energy at and above the critical density.⁹³ The energy thus absorbed at the surface is then transported into the bulk region via electron thermal conduction^{94,95} by a supersonic thermal wave until expansion of the surface plasma becomes appreciable and the resultant rarefaction or shock wave overtakes the thermal wave.¹³ The thermal transport rate or the speed of the thermal wave is normally

smaller than the thermal speed V_{te} of individual electrons (e.g. $V_{te} \approx 2.6 \times 10^8 \text{ cm/s}$ for electron temperature $T_e \approx 40 \text{ eV}$). This is because the hot electrons undergo many collisions in a random manner on their way to the colder region of high density of atoms and ions, thus reducing the speed of heat transport as the total thermal energy is summed over the Maxwellian distribution of the heat carrier electrons. Another negative effect on the rate of thermal transport is the plasma expansion, which takes heat away from being transported into the bulk region to accelerate the heavy ions.

FIG.6.1: Simultaneous scans of single-shot reflection (open circles) and transmission (closed circles) of the frontside probe are measured as functions of time delay, showing the characteristic time constants for plasma expansion(see text). Experimental conditions: $300\text{\AA}$ carbon-coated targets, femtosecond laser peak intensity = $5 \times 10^{14} \text{ W/cm}^2$.

For completeness of this discussion, first, to experimentally illustrate how fast the plasma expansion is at the front side of the target, Fig.6.1, similar to Fig.5.1a, is reprinted here to show the observed characteristic time constants of the plasma expansion in coated

fused quartz targets. These data, taken with the femtosecond laser, are single shot measurements of reflectivity (closed circles) and transmissivity (open circles) taken as functions of delay between the pump and probe pulses. The experimental procedures and set-up are the same as that described previously in Chapter 2, but now the target is carbon-coated pure fused quartz. In both data scans, the plateaus during early or negative delay correspond to the values for the cold target where the probe arrives prior to the pump and sees the target undisturbed by the pump. The cold undisturbed target absorptivity of $\approx 14\%$ is inferred from 7.8% reflectivity and 78% transmissivity that are measured with the probe beam. At some later delay when an overdense or super-critical plasma layer is formed at the surface during the onset of the pump pulse, the probe transmission quickly drops to zero. At the same time, the probe reflection rises rapidly. The 10-90% risetime of the reflection enhancement is about 1ps longer than the falltime of the transmission data. The reason for this is that, because of the absorptive film, an overdense plasma layer is produced much earlier during the leading edge of the pump pulse, thus quickly stopping transmission of the probe light. As shown in Chapter 5, the threshold for critical plasma formation is about $4 \times 10^{11} \text{ W/cm}^2$. This intensity level is at about 0.3ps prior to the peak of the pump pulse. This implies that, although the laser pulse is only 0.1ps at its full width half maximum (FWHM), the critical density plasma layer, where the probe transmission begins to turn off and reflection starts to increase, occurs at negative delay = -0.3ps, early during the rising edge of the pump pulse. Thus, during the subsequent time interval of 1ps, laser energy continues to be delivered at the surface plasma on the arrival of both the peak and the trailing edge of the pump pulse, making the plasma hotter, denser and more reflective as is observed by the reflection increase. Following the increase, the probe reflection experiences a much slower decay which is due to increasing absorption by the underdense or sub-critical plasma region as the overdense surface plasma cools and expands into the surrounding vacuum (see Chapter 4). The $1/e$ falltime for the reflection is $\approx 6\text{ps}$, determining the time scale for

appreciable plasma expansion. Whereas, the reflection risetime of 1ps signifies the time it takes for initiation of the plasma expansion. Because the heavy mass m_i of the glass ions is typically about 4×10^4 times larger than that of electrons and laser energy initially is delivered to heating of the light electrons, it takes as many subsequent collisions between ions and electrons as m_i/m_e to equilibrate the temperatures of electrons and ions. When ions gain sufficient energy from the energetic electrons, the plasma begins to expand. The plasma temperature of about 40eV, as was inferred from frontside probe frequency shifts, is related to the plasma expansion and corresponds to a maximum sound velocity of 4×10^6 cm/s (Chapter 4).

Now having experimentally determined the temperature of the overdense surface plasma and the important time scales for its subsequent expansion, we now turn to the results obtained from backside probing which is more interesting and is the topic of this chapter. Figure 6.2 displays the reflection (open circles) and transmission (closed circles) data for the backside probe with the same target. The data again show a quick drop in transmission and increase in reflection resulting from the rapid formation of a super-critical surface plasma, similar to that of frontside probe data. The strong enhancement of backside reflection shows that the plasma seen from the rear side also acts like a highly reflective mirror. The plasma gradient thus is initially rather steep, and its density must be much higher than the critical density to be so reflective. The reflection increase however is followed by a much slower decay time of 15ps than for the frontside probe reflection data, about 2.5 times longer than that of the slow decay in the frontside probe reflected signals (Fig.6.1) that are due to absorption by the expanding plasma at the front surface. It should be noted that during this 15ps decay, the pump laser heating has ceased and that the backside probe is not affected by plasma expansion as evidenced by the zero probe transmission data. These observations therefore lead to the realization that electron thermal transport phenomenon plays a dominant role in creation of the plasma behind the

surface. Note that the maximum backside reflectivity of about 42% is nearly twice larger than that of the maximum frontside reflectivity, indicating that the backside probe light suffers little absorption, if at all, by the expanding plasma at the front surface.

FIG.6.2: Simultaneous scans of single-shot reflection (open circles) and transmission (closed circles) of the backside probe are measured as functions of time delay. The solid line resulted from calculation discussed in text. Experimental conditions: 300Å carbon-coated targets, femtosecond laser peak intensity = $5 \times 10^{14} \text{ W/cm}^2$.

In Fig.6.3 the measured wavelength shifts $\Delta\lambda$ (solid dots) of the center-of-gravity wavelength component in the spectrum of the reflected backside probe are shown again as a function of delay. Each data point is an average of 90 laser shots and has a statistical uncertainty of $\pm 0.8 \text{ Å}$. Note that the measured shifts are observably large during the first picosecond of the reflection enhancement (Fig.6.2) and during which plasma expansion is negligibly small as evidenced by the frontside probe data. The shifts $\Delta\lambda$ are proportional to the velocity V_f of the steeply reflecting plasma surface or ionization front and are given by the Doppler formula:

$$\frac{\Delta\lambda}{\lambda} = -2n_{FQ} \cos \theta \frac{V_f}{c} \quad (6.1)$$

The factor of 2 accounts for reflection, $\theta = 19^\circ$, $n_{\text{FQ}} = 1.46$ is the fused quartz refractive index, c is the speed of light. The observed maximum shift of nearly $10\text{\AA}$ corresponds to a maximum velocity of $1.8 \times 10^7 \text{ cm/s}$. This is 4.5 times greater than the maximum ion sound velocity of the plasma as was inferred from the measured maximum frequency shifts of $7\text{\AA}$ in the frontside probe light shown in Chapter 4. Hence, the ionization front is supersonic and is attributed to a thermal wave that is created by electron thermal transport. It should be emphasized that the 1ps interval of the observedly large shifts corresponds to how fast the velocity of the moving mirror-like ionization front is. Whereas, the slow decay of reflection increase is an indicator of how long the plasma rear surface remains steep and hence specularly reflective. As will be seen below, this prolonged reflection decay is a measure of the lifetime of the plasma where plasma expansion has reached its full extent.

FIG.6.3: Observed wavelength shifts (circles) in the enhanced reflected backside probe light are large during the first picosecond after the pump pulse. Solid curve is time dependent shifts from the calculation from the nonlinear heat wave model.

6.3 Non-Linear Heat Wave Model For Plasma Formation

We now calculate the temperature and electron density evolution in the transparent region of the bulk material during the first few picoseconds where plasma expansion is negligibly small as was seen in the frontside probe data (Fig.6.1). In the coated targets the level of the transmitted pump light prior to formation of a critical plasma layer is much smaller than that in the uncoated target; and essentially no underdense bulk plasma is produced in the same manner as described in Chapter 5. Thus, an important role of the film is to promote large laser absorption at the surface, so that an overdense surface plasma layer is created much earlier prior to the peak of the pulse. This is to avoid an otherwise highly absorptive underdense bulk plasma that returns no probe reflection signals, and to allow more laser energy during the remainder of the pulse to be deposited at the surface plasma. As an overdense plasma layer is formed in the thin carbon region, it stops further transmission of the pump light and shields the remainder of the pump pulse from interacting with the bulk region behind the surface. The remainder of the pulse sends its energy into the surface region near the critical density, and the temperature in the carbon region at the surface is increased as rapidly as the pump pulse. This effectively confines laser energy deposition to the surface and sets up a high temperature ($\sim 40\text{eV}$) heat source at the surface for subsequent generation and propagation of a thermal wave, supported by electron thermal conduction, into the transparent bulk region. The generation of the thermal wave takes place during and after the laser heating by the pump pulse. As the thermal wave subsequently moves inward, it thermally ionizes the transparent material, giving rise to an ionization front that traverses the transparent region with a velocity equal to the rate of electron thermal transport. The electron temperature profile T_e and density N_e are governed by a nonlinear heat diffusion equation for thermal

electrons (in cgs units unless where specified), assuming no plasma expansion or zero ion temperature:

$$\frac{\partial}{\partial t} \varepsilon(x, t) = \frac{\partial}{\partial t} \left[\frac{3}{2} N_e T_e + \frac{1}{3} \left(2 \sum_{z=1}^8 N_z \chi_z + \sum_{z=1}^{14} N_z \chi_z \right) \right] = \frac{\partial}{\partial x} \left(k_e \frac{\partial T_e}{\partial x} \right) \quad (6.2a)$$

$$N_e = \sum_{z=1}^8 N_z^O z + \sum_{z=1}^{14} N_z^{Si} z \quad (6.2b)$$

The sum in the bracket is the total internal energy density ε . Here χ_z and N_z are the density and ionization energy of ions with atomic number z . The first term is electron thermal energy, and the second and third terms are energies stored in the plasma ionization states of oxygen and silicon ions, respectively. The term in the right hand side is the electron energy flux with Spitzer-type electron thermal conductivity,⁹⁶ i.e. $k_e = k_o T_e^{5/2} / Z_{eff} \approx 2 \times 10^{-4} T_e^{5/2} / Z_{av}$, where $Z_{av} = N_e / N_a$. The left boundary condition to the equation is taken as ⁹⁷ $T_e(0, t) = T_o e^{2\gamma}$ for $t \leq 0$, and $\partial T_e(0, t) / \partial x = 0$ for $t > 0$. The right boundary condition is $T_e(+\infty, t) = T_{room} \approx 0eV$, signifying the finite extent of the thermal heat wave. The temperature at the surface ($x=0$) rises exponentially and as quickly as the laser pulse from the room temperature to a maximum value T_o which is determined by the total absorbed energy of the pump pulse. Pump laser energy absorption of approximately 56% is inferred from the 36% specular reflection and 8% transmission of the pump pulse. The level of diffusely scattered light of the pump pulse has been measured by others ^{16,72} and is small (<5%) because during the short pump pulse there is very little plasma expansion and the target surface remains optically flat. After the heating ($t > 0$) no plasma expansion or no heat flux at the surface into the vacuum is allowed as described by the left boundary condition for $t > 0$.

Seemingly intractable indeed, the above equation when coupled with the LTE model described by the Saha equations (Chapter 4) for temperature dependent ionization states of variously ionized atoms poses a challenge for analytical solutions and numerical as well. Instead of following this approach, I now discuss an approximation to reduce these coupled equations to a single nonlinear but solvable differential equation: The ionization energies I_n is treated as a continuous function of ionization states n of ions as was shown by Eqn.(4.4) in Chapter 4. The Saha equations, such as Eqn.(4.3), determines the ion density N_n as a function of electron temperature. Using these equations and the calculation method discussed therein, the terms in the brackets on the left side of Eqn.(6.2) can be expressed by a best fit function of electron temperature in the temperature range (0-10⁶K) or (0-100eV), i.e.

$$\varepsilon(x,t) = \frac{3}{2} N_e T_e + \frac{1}{3} \left(2 \sum_{i=1}^8 N_i^O \chi_i + \sum_{i=1}^{14} N_i^{Si} \chi_i \right) \approx a T_e^{1+\alpha} \quad (6.3)$$

where $\alpha = 1.5$, and the proportionality constant $a = 1.4/(\text{deg})^{(1+\alpha)}$. Figure 6.4 displays the LTE calculation results (dots) and the best fit curve described by Eqn.(6.3). Using this approximation, Eqn.(6.2) can be rewritten simpler in terms of electron internal energy, i.e.

$$\frac{\partial \varepsilon(x,t)}{\partial t} = k_c \frac{\partial}{\partial x} \left(\varepsilon^q \frac{\partial}{\partial x} \varepsilon \right) \quad (6.4)$$

where $k_c = k_0/(1+\alpha)a^{1/(1+\alpha)}$, $q=(5/2-\alpha)/(1+\alpha)$. This equation is subject to conditions similar to the above conditions and the energy conservation with total energy fluence absorbed at the target surface F_a , i.e.

$$\int_0^\infty \varepsilon(x,0)dx = F_a \quad (6.5)$$

FIG.6.4: LTE calculation (dots) of internal energy as a function of temperature in the range 0-10⁶K. Solid curve is the best fit curve described by Eqn.(6.3).

Equation (6.4), describing propagation of a thermal energy wave, does not take into account radiation transport and contribution of ions to thermal transport process. First, to test whether x-ray emission and its effect are significant, we compare the backside probe reflection data between coated pure fused quartz (SiO₂) targets (Fig.6.2) and coated glass

targets. The glass targets contain 27% impurities of readily-ionized materials and 73% SiO_2 ⁴⁸ (Fig.5.4, Chapter 5). As can be seen, the coated glass reflection decay time of about 1.5ps is 10 times faster than that of the coated pure fused quartz. Also, since x-ray photons of energies several times greater than 40eV emitted from the plasma of 40eV will dominate in the radiation transfer process in the plasma,¹³ the valence electrons are easily excited by such photons into the conduction bands with the band gap energy of 9eV. Thus, had the x-ray emission been dominant or had sufficiently strong flux from the surface plasma into the bulk region, we would have expected no significant difference in the reflection data for both targets, regardless of the impurities. Furthermore, total Bremsstrahlung emission accounts for less than 1% of the plasma energy;⁹⁸ and the total fluence of blackbody radiation emitted from the surface plasma of about 40eV is negligibly small ($0.3\text{J}/\text{cm}^2$), compared to the absorbed laser intensity ($25\text{J}/\text{cm}^2$). This therefore verifies conclusively the insignificance of x-ray preheating of the bulk material. The large difference observed in the reflection decay times between targets of glass with impurities and pure quartz are merely attributed to the presence of the impurities.⁴⁸ The impurity atoms in the glass target are readily ionized by collisions with energetic electrons and high field ionization, giving rise to a largely absorptive underdense plasma embedded in the bulk region that leads to a faster reflection decay in the glass target.

In the above equations, heat transport by ions is also omitted because its large mass is $\sim 4 \times 10^4$ times larger than that of electrons; and the ionic thermal conductivity is reduced by a factor $\sqrt{(m_e/m_i)} = 2 \times 10^{-2}$.⁹⁹ Another assumption of the model is the use of the Saha equations which may not be completely valid because of the short time scale of interest. However, under the current conditions, the Saha equations should be a good approximation for several following reasons:⁷⁵ Collisional rates in high density are predominant over the radiative processes; the electron-electron equalization time is $< 10\text{fs}$, much shorter compared with the cooling time by expansion and therefore quickly

resulting in a Maxwellian distribution of electrons; and finally high stage ionization rates are small for temperature $< 100\text{eV}$. Another assumption made is the use of Spitzer-type thermal conductivity for electrons which may not be valid for high temperature and solid density plasmas. Comparison between the Spitzer conductivity and many other models has been made^{100,101} and shown some unsolved uncertainty as to what model gives the most reliable predictions on the thermal properties of the high temperature and dense plasmas. This is one of the many pure scientific issues and remains a challenge to theorists to better understand the fundamental thermal properties of such hot and dense matter. However, the Spitzer-type conductivity is generally thought to be a good approximation and often used for plasmas of temperature greater than 10eV . For example, the Lee and More (LM) numerical model, which is often thought to be more "realistic", shows that for solid density plasmas Spitzer conductivity results in a good agreement with LM model for plasma temperature $> 12\text{eV}$.¹⁰⁰

The approximate analytical solution to the thermal heat wave and its front position x_f for $t < 0$ is similar to that of the Marshak radiation wave²⁹ and for $t > 0$ the same as that of Zeldovich¹³ (see Appendix A for more algebraic details):

$$\varepsilon(x, t_s) = \varepsilon_o \text{Exp}(2\gamma) \left(1 - \frac{x}{x_f(t)}\right)^{1/q} \quad \text{and} \quad x_f(t_s) = \frac{1}{q} \sqrt{k_c \frac{\varepsilon_o^q}{\gamma}} \text{Exp}(q\gamma) \quad (6.6a)$$

and

$$\varepsilon(x, t_s) = \left(\frac{F_a^2}{k_c(t + t_{mat})} \right)^{\frac{1}{2+q}} \left(1 - \frac{x^2}{x_f^2(t)}\right)^{1/q} \quad \text{and} \quad x_f(t_s) = \xi_o \left[k_c F_a^q (t + t_{mat}) \right]^{\frac{1}{2+q}} \quad (6.6b)$$

In the above, ξ_0 is determined by Eqn.(6.5), and t_{mat} is the time to match the solutions before and after the laser heating, i.e. $x_f(t_{\leq}) = x_f(t_{>})$. Since the ion acoustic velocity is proportional to $\sqrt{T_e}$, from Eqns.(6.3) and (6.6) it can be seen that the plasma expansion velocity scales as $V_{\text{TP}} \sim t^{0.08}$. By contrast, the heat wave front velocity scales as $V_f = dx_f/dt \sim t^{-0.58}$. Clearly, at early time the heat wave front is supersonic and outruns the rarefaction wave, and the cooling of the surface plasma by thermal transport is more effective than hydrodynamic expansion.

It should be emphasized that at the time $t=0$ laser energy delivery to the target surface is stopped and cooling by predominant thermal transport into the colder region of the target begins. For this reason, Eqn.(6.4) accepts two different solutions which are due to the difference in the boundary conditions at $x=0$ as stated above. At the end of the laser heating, $t=0$, the target surface temperature reaches its maximum T_0 . At this time, as shown in Fig.6.5, the thermal wave front propagation distance or the plasma thickness is only $\approx 400\text{\AA}$, which is about the same order of magnitude of the plasma optical skin depth. Thus, at the time when laser heating ceases, the thickness of the plasma is equivalent to the depth of the absorption region which serves as a heat source for further thermal conduction. For this reason, at time $t=0$ the surface plasma of $400\text{\AA}$ thickness is used as a source at the target (boundary) surface for subsequent propagation and cooling of the thermal wave. Another point worthwhile mentioned is that details of the temperature or density profiles in the region above the critical density level do not significantly affect the backside probe light and its reflected signals. This is as will be shown below due to the existence of an optimal density where (probe) laser light is maximally absorbed and the laser electric field is minimally vanishing.

6.4 Simulation and Analysis

FIG.6.5: Calculated electron temperature profiles at different instants as predicted by the nonlinear heat wave model.

FIG.6.6: Calculated electron density profiles at different instants using the simple approximate method to the Saha equations.⁷⁶

Figure 6.5 displays calculated snapshots of the temperature distribution at different instants. The surface temperature from room temperature rises rapidly, reaching to a maximum value of $\approx 40\text{eV}$. At the same time, the thermal wave front moves inward supersonically. For $t > 0$, the surface temperature begins to relax gradually; and the

thermal wave slows down as diffusion of the absorbed energy continues further into the underlying colder region to ionize the transparent material. Figure 6.6 shows the corresponding electron density profiles normalized to the critical density N_{cr} . As is expected, during the pump pulse the super-critical surface plasma as seen from the back side progressively becomes denser and thicker as a result of thermal transport, making the plasma more reflective as was observed. At later time, the plasma cools and its scale length in the bulk region increases, leading to increasing absorption of the probe light. This hence results in the observed long decay in the backside probe reflection. Had the heat diffusion not been important, the plasma back surface would remain highly reflective for as long as the plasma's lifetime of many picoseconds. Moreover, if this were the case, the backside reflectivity of the surface plasma would remain as constant and high as the reflectivity calculation without absorptive effects of a bulk plasma demonstrated in Fig.5.1a (Chapter 5). This thus further confirms the importance of the electron thermal transport mechanism which is necessary to account for the backside reflectivity slow decay. The plasma lifetime, t_p , is approximately determined by the distance of a rarefaction wave which is resulted from plasma expansion and travels with the expansion velocity V_{TP} into the heated material (see Chapter 4). When the edge of the rarefaction wave reaches and overtakes the heat wave front, electron thermal transport still continues but now in the material that expands into the opposite direction, i.e. when

$$x_f(t_p) = \int_0^{t_p} V_{TP}(t') dt' \quad (6.7)$$

This relation yields the lifetime of $t_p = 24.4$ ps, where the thermal heat and rarefaction waves both have traveled and met at a distance of 5350Å inside the bulk material. It turns out this lifetime is equal to the full duration of the backside reflection decay (Fig.6.2). As one would expect, at the end of the reflection decay when the rarefaction wave front has fully depleted the plasma in the bulk region, most of the plasma produced by the heat

wave front has undergone expansion and cooling, leaving the target surface annealed with a large underdense plasma at the front. As a result, the observed backside reflectivity of about 4% becomes similar to that of Fresnel reflectivity of the fused quartz interface, and the transmission remains zero due to large absorption by the underdense plasma. The penetration depth of the heat wave or rarefaction wave is also the maximum thickness of the plasma which determines the amount of the materials needed for total plasma development.

The velocity of the moving mirror-like plasma surface, as seen from the rear, varies as a function of time, and thus different portions of the finite probe pulse suffer different amounts of blue shifts. In addition, the frequency shifts shown in Fig.6.3 are detected during the early delay where reflection enhancement is strongest and the plasma reflectivity remains approximately constant. For these reasons, any spectral changes due to amplitude modulation of the probe light can be neglected. Figure 6.3 also displays the calculated profile (solid curve) of the velocity or wavelength shifts convolved over the probe pulse, showing fairly good agreement with the experimental results at time $< 0.7\text{ps}$. At late delay $\geq 0.7\text{ps}$, the heat wave front velocity slows down considerably and becomes more difficult to measure accurately. The discrepancy in the data at late delay $t_d \geq 0.7\text{ps}$ suggests some reduction in the rate of the electron thermal transport. This reduction is attributed to three possibilities:

1. The combined effect of the thermal transport and appearance of a rarefaction wave resulting from plasma expansion. The rarefaction wave tends to carry energy in the opposite direction to the thermal wave, giving rise to a decrease in the rate of thermal transport into the bulk region. The reduction suggests inhibition of electron thermal transport which has been reportedly observed in the past.¹⁰²⁻¹⁰⁵ Although there have been conflicting reports as to whether electron thermal transport is strongly

inhibited;^{102,106-108} and much unresolved confusion over the specific mechanisms for the inhibition,¹⁰⁹ it is however reasonable to generalize that the inhibition of thermal transport is mainly due to the plasma expansion. Hydrodynamic expansion results in a host of many nonlinear processes, such as ion acoustic turbulence, electrostatic or dc magnetic fields,³¹ that have been suggested as mechanisms for thermal inhibition.¹⁰⁹

2. At delay times ≥ 0.7 ps when the spatially averaged temperature of the plasma as shown in Fig.6.2a is ≤ 12 eV, the true thermal conductivity of the plasma may be less than that predicted by the Spitzer-type conductivity and thus results in much smaller frequency shifts. To fully account for strongly-coupled plasmas, effects of Fermi electron degeneracy, electron-ion and ion-ion correlation, and quantum effects must be considered. Lee and More's calculation¹⁰⁰ shows that for solid density plasmas with temperature ≤ 12 eV Spitzer conductivity may not be used because of the above-mentioned effects which have been omitted in the Spitzer formulation. And finally,

3. The above simple diffusion equation may be no longer valid for the plasma of sharp temperature gradients as shown in Fig.6.5. The ratio of electron mean free path to the temperature scalelength, $\lambda_e/L_T \sim 0.1$, is much greater than the prescribed upper limit of 2×10^{-3} .¹¹⁰ Thus, it may require nonlocal kinetic effects due to fast unthermalized electrons to account for the reduction.¹¹¹

Having determined the plasma temperature and electron density evolution from the nonlinear heat wave model, we now simulate the reflection and transmission of the backside probe light interacting with such plasma as functions of time delay between the pump and probe pulses. As for the probe transmission, it can be easily seen that the transmissivity is similar to the calculation demonstrated in Fig.5.1b (Chapter 5) where rapid formation of the overdense surface plasma quickly turns off the probe transmission.

Because of the carbon thin film, calculation of the probe reflection is somewhat complicated. At early delay, $t_d \leq t_{cr} = -0.3\text{ps}$, prior to formation of a critical plasma at the surface, the carbon film is lightly heated and its optical properties suffers little or essentially no changes. At later times $t_{cr} < t_d < 0$, the overdense plasma layer of $\leq 300\text{\AA}$ thickness is created within the carbon region. The probe reflection occurs at both the plasma-carbon and carbon-quartz interfaces. We therefore can treat the thin surface plasma as a normal thin film problem and use Fresnel formula for calculation of reflectivity.^{71,112} The thin film calculation for $t_d < 0$ uses an dielectric function as following, taken into account the contributions of covalent electrons and plasma in the carbon film:⁵⁹

$$\epsilon \approx \epsilon_c \quad \text{for } t_d \leq t_{cr}$$

$$\epsilon = \epsilon_c - \frac{N_e / N_{cr}}{1 + i\nu_{ei} / \omega} \quad \text{for } 0 > t_d \geq t_{cr} \quad (6.8)$$

where $\epsilon_c \approx 2.5 + 0.95i$ is the complex dielectric function of the amorphous carbon film,¹¹³ and ν_{ei} is the electron-ion collision frequency. The reflectivity of the thin film-like plasma for S-polarized backside probe light is given by:

$$R \approx \left| \frac{\sin(\theta_{inc} - \theta_{int})}{\sin(\theta_{inc} + \theta_{int})} \right|^2 \quad (6.9)$$

In the above, $\theta_{inc}=19^\circ$ is the angle of incidence at the front surface, and θ_{int} is the complex angle of the probe light propagation inside the plasma and is given by the generalized Snell's law of refraction with $\theta_{int}=\arcsin\{\sin(\theta_{inc})/n_r\}$ and $n_r=\text{Re}(\sqrt{\epsilon_c})$.

As shown in Fig.6.6a, after the laser heating where the heat wave front goes far beyond the carbon film region into the transparent fused quartz region, the probe radiation from the rear side is attenuated quickly within the optical skin depth at N_{cr} and can not reach the region of the carbon film at the front side. Consequently, the probe reflection takes place only at the plasma-quartz interface. Calculation of the probe reflectivity of the plasma after the laser heating is determined from the Helmholtz equation:

$$\frac{d^2 E}{dx^2} + k^2(\epsilon_p - \sin^2 \theta_{inc})E = 0 \quad \text{for } t_d \geq 0 \quad (6.10)$$

In the above, E describes variation of the S-polarized probe electric field along the x -axis direction, and k is the wave number in the vacuum. ϵ_p is the complex dielectric function of the plasma sensed by the probe light from the rear and is given by

$$\epsilon_p = \epsilon_{FQ} + i \frac{4\pi\sigma}{\omega} = \epsilon_{FQ} - \frac{N_a / N_{cr}}{1 - (N_a / N_{cr}) / \omega} \quad (6.11)$$

with

$$\epsilon_{FQ} = \frac{1 + 8\pi\alpha(N_a - N_i)/3}{1 - 4\pi\alpha(N_a - N_i)/3} \quad (6.12)$$

σ is the plasma high frequency electrical conductivity. N_a is the number density of the fused quartz (FQ) material, and N_i is the density of the ionized atoms. ϵ_{FQ} is the dielectric function due to $(N_a - N_i)$ neutral atoms or covalent electrons with $\alpha = 2.94 \times 10^{-24} \text{cm}^3$ as the mean atomic polarizability due to bound electrons. Eqn.(6.12) basically is the Lorentz-Lorenz formula,¹¹² used to account for un-ionized atoms near the mixed region of plasma-FQ interface. When there is no ionized atom in the region, the formula

yields a value $n_{FQ} = 1.46$ for the refractive index of the FQ materials.⁴⁹ When all atoms are ionized, $N_i = N_a$, the dielectric function corresponds to the usual Drude's model for plasmas.¹¹⁴

Calculation of the plasma reflectivity using Eqns.(6.8-12) requires detailed description of the plasma collisionality. The plasma collisionality is the most important and fundamental plasma parameter that dictates the laser-plasma interaction, absorption, transport mechanisms, and many other subsequent processes as functions of time and space. Recently, there have been various models, including parametric and quantum mechanical models, proposed for the plasma collisionality in attempts to describe and predict short pulse laser light absorption in solid and important plasma parameters.^{10,51,71,95,100,101,115-117} However, these models, based on certain assumptions, are not complete enough to reflect all important aspects and dependencies of the plasma collisionality. They often underestimate effects of strongly coupled interaction between the neighboring charged particles and can not reconcile important differences in the physics of the normal and anomalous laser absorption mechanisms that are strongly regulated by the plasma collisionality. Further discussion of the discrepancies and weaknesses of these models is beyond the scope of this research; and to be sure, when a satisfactory model for plasma collisionality is attained, much new and interesting fundamental physics of high temperature and density plasmas will be revealed.

Nevertheless, in the spirit of this experimental work and keeping theoretical analysis simple for the purpose of the current calculation, we can reasonably employ a parametric model, which has been used previously by many researchers to account for Milchberg's observation of resistivity saturation in hot solid density plasma:^{10,17,116,118}

$$v_{ei, sat.} = v^* \frac{N_e}{N_{cr}} \frac{[0.1T_e^2 / Z_{eff} + 1]}{[0.1T_e^2 / Z_{eff} + (N_e / N_{cr})^{2/3}]} \quad (6.13)$$

where v^* is the collision frequency at the critical density, N_{cr} , and will be treated as a fitting parameter. Z_{eff} is the effective charge state of the plasma, and T_e is electron temperature in eV. Many important physical features of this model can be realized from the Eqn.(6.13). First, apart from the weak density dependence in the bracket, the plasma collisionality varies linearly with the plasma density, asymptotically similar to that of Spitzer-type collisionality which is valid for high temperature and low density plasmas. In low temperature and high density regime, the collision frequency is proportional to $N_e^{1/3}$ which is as expected. In this regime, solid-state effects become important, and the ion-electron scattering length is limited by the average spatial interionic separation as a minimum mean free path between two consecutive scatterings¹⁷ since the electron minimum scattering length between two electron-atom collision can not be greater than the interionic distance. The collision frequency is approximately given by the ratio of the electron thermal velocity V_{te} and the interionic spacing R_a , i.e.

$$v_{ei, mfp} \approx \frac{V_{te}}{R_a} = \frac{(k_B T_e / m_e)^{0.5}}{(4\pi N_i / 3)^{-1/3}} \sim N_e^{1/3} \quad (6.14)$$

It should be mentioned here that the interionic spacing at solid density is only 1.58Å, about 3 times smaller than the atomic orbital radius (4-7Å) of the outermost electrons in silicon atoms. This thus leads to strong effects of electron-electron interaction and electron degeneracy. Equation (6.13) also includes effects of Fermi electron degeneracy in the high density plasma. Since collisions involve only electrons near the Fermi surface and the Fermi energy of these electrons is $\epsilon_F \sim N_e^{2/3}$,¹¹⁹ the saturation model can be reduced to the empirical formula that was used by several researchers to obtain the best fit Milchberg's observation of resistivity saturation.^{17,118}

Now having set up the formulation for the interaction between the plasma and backside probe light, we solve the Helmholtz equation for reflection and absorption of the probe radiation as following. In the interior region far from the plasma, the probe electric field can be written in the form:

$$E = E_i \exp(-ikx - i\omega t) + E_r \exp(+ikx - i\omega t) \quad (6.15)$$

where E_i and E_r denote the electric field amplitude of the incident and reflected waves, respectively. The reflectivity of the plasma is given by:

$$R = \frac{|E_r|^2}{|E_i|^2} \quad (6.16)$$

Propagation and local absorption of the probe light are described in the plasma region and is given by Poynting's theorem:¹¹⁴

$$\frac{\partial \xi}{\partial t} = \langle \text{Re}(j) \text{Re}(E) \rangle = \frac{1}{2} \text{Re}(\sigma) |E|^2 = \frac{\nu_{ei}(x) N_e(x) / N_{cr}}{1 + \nu_{ei}^2(x) / \omega^2} \frac{|E(x)|^2}{8\pi} \quad (6.17)$$

where j is the charge current. The left hand side denotes the rate of the probe laser energy deposited in a unit volume of the plasma. It can be easily seen from Eqn.(6.17) that there exists a density level at which energy deposition is maximized. In the interior of the target where N_e/N_{cr} , $\nu_{ei}/\omega \ll 1$, propagation of the probe laser light is strongly oscillatory and unattenuated. By contrast, as it travels further inside of the plasma region where N_e/N_{cr} , $\nu_{ei}/\omega \gg 1$, the field is heavily absorbed and attenuated over the optical skin depth of the plasma. Thus, there is a competition between the term $|E(x)|^2$ and $\nu_{ei}(x)N_e(x)$ in Eqn.(6.17) which makes it maximized locally at some optimal density N_{op} , giving rise to localized absorption. Another reason for examining this optimal density is that details of the plasma density profile in the region $\gg N_{op}$ are insensitive to the calculation of the

field E and can be safely ignored because the field there is rapidly evanescent and vanishing. Hence, calculation of the plasma reflectivity in the bulk target far from the plasma would be simplified and sensitive only to the shape of the density profile from and below the N_{op} level. Also, since the details of the density profile above N_{op} and nearer to the target surface are not seen by the backside probe light, effects of a rarefaction wave will not complicate the probe reflected signals.

FIG.6.7: Time variations of parametric collisionality that yields best fits to the reflectivity data (curve a), and of temperature at optimal density (curve b).

The solid curve in Fig.6.2 is resulted from numerical integration of the wave equation (6.10) by the standard Runge-Kutta method,¹²⁰ up to the lifetime of the plasma $t_p=24.4\text{ps}$. At early delay time $\leq -0.3\text{ps}$, where the plasma thickness is $\leq 300\text{\AA}$ and is formed within the region of the carbon film, the probe reflection occurs both at the plasma-carbon and carbon-quartz interfaces, the reflectivity is similar to that of thin film Fresnel-like. At later times, when subsequent the thermal wave strongly moves into the quartz region and ionizes the neutral atoms, only a single reflection from plasma-quartz interface occurs. Calculation of reflectivity is performed with v^* as the only adjustable

parameter for best fits to the data. Curve (a) in Fig.6.7 shows the values of v^* normalized to the laser frequency ω at different time delays. The lower and upper limits of v^*/ω are 0.29 and 0.55, respectively, which are about the same order of magnitude as often parametrized previously by many researchers.^{10,51,72} Figure 6.7 also displays the corresponding time variation of the plasma temperature at the optimal density, showing that v^* is a decreasing function of temperature. This is as expected since the classical Spitzer-type collision frequency is also a decreasing function of temperature.

FIG.6.8: Time variations of collisionality ratio between the values by saturation model and mean free path (curve a), and values of calculated collisionality (curve b).

In Fig.6.8, we show the ratio of the calculated collisionality to that determined by the electron minimum mean free path, indicating that the collisionality is less than and close to that predicted by the minimum mean free path related values. This is suggestive of the strong effects of electron degeneracy and inter-particle correlation at solid density. Curve (b) in the figure shows the values of collision frequency calculated at the optimal density and reveals that fact that the earlier the delay where the density profile still remains steep and thin ($L/\lambda \ll 1$), the maximum laser absorption takes place at the higher density.

The time dependence of the optimal density normalized to the critical density (curve a) is shown in Fig.6.9, indicating that at early time, where the plasma rear surface gradient is rather steep or the scalelength is short, $L/\lambda \ll 1$, most of the laser energy is delivered to the overdense region much above N_{cr} .^{72,118} This, together with detailed understanding of the collisionality and laser pulse shape or rate of laser energy delivery, will enable us to design solid materials as an effective absorber which is currently a topic of great interest in obtaining ultrafast bursts of short wavelength radiation.⁹ Curve (b) in the figure shows the time development of the plasma scalelength L due to electron thermal transport. The scalelength is approximately obtained when fitting the plasma profiles in the interior region shown in Fig.6.6 to a linear form, $N_e(x,t) = N_e(0,t) \{1 - x/L\}$. In Fig.6.10, the optimal density is plotted against the plasma scalelength L/λ . The circles in decreasing order correspond to values at $t_d = 0, 5, 10, 15$, and 20 ps, respectively. The solid curve is the best fit and scales the optimal density as $N_{op} \sim \sqrt{\lambda/L}$, which is consistent with previous works by other researchers.^{51,118}

FIG.6.9: Time dependence of calculated optimal density and scalelength.

FIG.6.10: Optimal density, where laser deposition is maximum, varies a function of the plasma scalelength.

6.5 Summary and Implications

In this chapter, I have shown that in probing electron thermal transport the difficulties realized from the previous works presented in Chapters 3, 4 and 5 can be successfully avoided. These include the nonlinearity in the refractive index of the transparent target, production of an overdense surface plasma and its subsequent expansion that limit the accessibility and complicate signals of a frontside probe light, and formation of a large underdense plasma in the bulk target which returns no information on transport mechanisms. Better understanding of these difficulties together with the realization that any changes in the material properties of the bulk region, without any hydrodynamic effects, are clear signature of the important energy transport mechanisms, had led us to use pure fused quartz targets overcoated with a carbon film. The film is highly absorptive, so that it reduces the plasma formation threshold and transmitted pump energy level to eliminate the above-mentioned difficulties. In addition, it enhances laser absorption and

confines laser energy deposition to the surface, allowing a high temperature heat source to be established for subsequent generation and propagation of a thermal wave, supported by electron thermal conduction, into the underlying cold region behind the target surface.

We have seen experimental results from frontside and backside probing which reflect experimentally the totality of plasma evolution into both sides of the target surface. The frontside probe data determine the characteristic time constants for rapid formation and subsequent expansion of a super-critical surface plasma into the surrounding vacuum. The observed backside probe reflection enhancement indicates that the plasma has a steep gradient and its steepness, as is seen from the rear, persists for a long period approximately equal to the lifetime of the plasma. The measured backside probe frequency shifts further confirm that the ionization front, created by electron thermal transport, acts like a highly reflective moving mirror and is supersonic during the time scale of ≈ 1 ps after laser irradiation. In the intensity regime and time scale of the current experimental conditions, experimental checks and arguments were made to conclusively justify the insignificance and hence omission of many other processes, such as x-ray emission and many nonlinear effects associated with plasma expansion. The backside probe data has demonstrated we can successfully isolate thermal transport from these complex effects.

A nonlinear heat wave model for electron thermal transport, combining the important features of Marshak wave during the laser heating and Zeldovich wave after laser heating, has been proposed. The model predicts the surface plasma evolution in both time and space that gives rise to the observed behavior in the experimental data. Calculations of frequency shifts using the model yield good agreement to the observations. This agreement suggests that electron thermal conduction is the most effective energy transport mechanism into the bulk solid during the first picosecond after laser irradiation.

It is the gradual heat conduction by electrons after the laser heating that accounts for the observed prolonged decay in the reflection data. Also, we have solved the Helmholtz wave equation for the probe electric field together with a proposed model for the plasma dielectric function to simulate the observed reflectivity. The proposed dielectric model includes both a contribution from the bound covalent electrons which is determined by the Lorentz-Lorenz relation and a contribution from free electrons that is equivalent to the usual Drude's model for plasmas. Also, the dielectric model concludes of a semiempirical collisionality to predict laser deposition and plasma absorptivity. The laser deposition is found to be locally maximized at some optimal density level. Time development of the optimal density allows us to ignore unnecessary details of plasma density profile above the optimal level where a rarefaction wave may have developed. This is to ensure simplification in the analysis of the backside probe frequency shifts and reflection. The collisionality was proposed to take into account solid-state effects and inter-particle correlation that have been omitted in the classical formulation. The proposed collisionality is the limiting case of both Spitzer-type collisionality for high temperature and low density plasmas and Lee and More's model for low temperature and high density plasmas. Finally, the numerical integration of the Helmholtz equation yields optimal density levels and the optimal density scales as $\sim\sqrt{\lambda}/L$, consistent with previous studies for short scalelength and high density plasma.

The experimental data suggest that transport of thermal electrons initially is not strongly inhibited; and it becomes reduced when plasma expansion, as often suggested in the past,¹⁰⁹ begins to set in. This thus opens the way for more reliable observation and careful experiments to verify whether there is strong inhibition in electron thermal transport. In addition, detailed analysis with hydrodynamic effects taken into account will help to characterize the degree of the inhibition. Another implication of this work involves the concept of nonlocality in electron thermal transport that might result in

thermal inhibition.¹¹¹ The concept, though not yet conclusively confirmed, originates from the fact that the energies of (super-thermal) electrons are so large that the electron mean free path between collisions becomes much greater than the local temperature gradient. The Fourier theorem for heat diffusion thus is no longer valid.¹²¹ Superthermal electrons tend to run away and travel a long distance, decoupling from nearby colder electrons and giving rise to nonlocal characteristics of the energy transport. For the intensities $<10^{16}\text{W/cm}^2$, however, the effects of the superthermal electrons are of little significance;⁹³ and the validity of the nonlocality concept has to await higher power lasers and spatially-resolved detection of electron temperature at various depths inside the target by line radiations emitted from the plasma,¹²² (see Chapter 8). On the issues of laser-matter coupling, the calculation presented above reflects that the plasma collisionality, as the most fundamental and important parameter, be further investigated. One reliable and rather complete approach, combined from previous works by other researchers,^{10,17,50,65,71,73,116,118,123} toward this investigation is time-resolved measurements of angular- and polarization-dependent reflection of both frontside and backside probes. Angular dependent reflectivity yields estimates of the plasma scalelength because the reflecting or turning point density scales as $N_{\text{TP}}=N_{\text{Cr}}\cos^2\theta$. The difference in P- and S-polarized probe reflection is used to infer laser absorption, and hence collisionality. Analysis of the frontside probe reflectivity will provide information about the plasma collisionality during laser heating of the target. On the other hand, the backside reflectivity yields collisionality information that accounts for heat transport phenomena in the bulk region after the laser heating. These measurements should further complement the current work. More importantly, they provide detailed and consistent understanding of the plasma collisionality which is really manifested through laser absorption and dictates many subsequent plasma behaviors.

Finally, the results found from this work determine the spatial extent of the plasma which relates to the maximum amount of the solid materials that can be heated to high temperature for high power energy conversion. This is an important issue in the current developments of x-ray laser or ultrashort x-ray sources by using high temperature and solid density plasmas.

CHAPTER 7

CHARACTERISTICS OF HIGH INTENSITY PULSE PROPAGATION

Laser pulses of sufficiently high intensity propagating in a transparent medium will be increasingly absorbed and attenuated because, as was shown in Chapter 5, the material absorptivity is exponentially proportional to the laser intensity. Absorption of laser photons is regulated by inverse Bremsstrahlung mechanisms, causing ionization of easily-ionized atoms of impurities in the propagation path of the laser pulse. At the same time, a negative change in the refractive index, corresponding to a positive change in the electron plasma density, results in a blue frequency shift in the transmitting laser light. In this chapter, measurements of the high intensity transmitted portion of the laser pump pulse in the pump-probe experiments with both uncoated and coated transparent targets, described in the previous chapters, are given. Characterization of the intensity thresholds and timings for induced phase modulation and formation of sub-critical and critical density plasmas are presented. Energy and spectral measurements of the high intensity transmitted (pump) light as functions of incident energy are discussed. These measurements yield estimates of ionization rates and effects of pulse temporal narrowing and spectral broadening.

7.1 Intensity Levels and Timings of Important Thresholds

FIG.7.1: Measured and calculated characteristic intensity thresholds during the incident Sech^2 laser pulse (solid curves) of 1.2ps FWHM (a): Induced phase modulation, I_{IPM} , sub-critical plasma formation, I_{sub} and critical plasma formation thresholds I_{cr} . Dashed curve is the profile of transmitted pulse. (b) is for 200fs FWHM laser pulses

As a laser pulse of 1.2ps FWHM (solid curve in Fig.7.1a) is incident onto a transparent target, early during the leading edge of the laser pulse where the intensity is below the sub-critical plasma density ($\sim 10^{18}/\text{cm}^3$) formation threshold, I_{sub} , the weak probe pulse in the pump-probe experiments discussed in Chapter 3 suffers red frequency shifts that are proportional to the pump laser intensity. The observed smallest red shift of $0.25\text{\AA}$ (see Chapter 3) corresponds to the intensity threshold, $I_{\text{IPM}} \sim 1 \times 10^{12} \text{W}/\text{cm}^2$, for observable effects of modulation in the medium refractive index induced by the strong pump pulse onto the probe light (induced phase modulation, IPM) or onto itself (self-phase modulation, SPM). The time $t_{\text{IPM}} = -1.3\text{ps}$ occurs during the leading edge of the pulse. Figure 7.1a also displays the calculated profile (dashed curves) of the transmitted portion of the pump pulse exiting at the rear surface of the target. As can be seen, initially prior to the time $t_{\text{sub}} = -0.6\text{ps}$ where a subcritical plasma of $\geq 10^{18}/\text{cm}^3$ begins to form at the surface there is very little absorption and $\approx 96\%$ transmission of the laser energy. During this time interval, $t_{\text{sub}} - t_{\text{IPM}}$, the phase modulation effects are predominant. Also, since the intensity is monotonically increasing, or $dI/dt > 0$ during this interval, the effects are frequency shifts and/or broadening toward the red side of the laser pulse spectrum. In Chapter 5, it was estimated that the subcritical plasma is formed at the surface when laser intensity at the surface reaches $I_{\text{sub}} = 9.6 \times 10^{12} \text{W}/\text{cm}^2$ and the critical density plasma at $I(t_{\text{cr}}) = 1.3 \times 10^{13} \text{W}/\text{cm}^2$. The critical time occurs at $t_{\text{cr}} = -0.46\text{ps}$ prior to the peak of the pulse. Figure 7.1b shows the corresponding intensity thresholds for the case of shorter laser pulses of 200fs FWHM. In either cases there is a sudden drop in the profiles of the transmitted pump pulse during the interval $(t_{\text{cr}} - t_{\text{sub}})$ when laser absorption by the bulk underdense plasma is predominant. This sudden decrease in the transmitted pulse profiles due to the rapid buildup of plasma density has been observed previously with laser pulse of 20ps duration.^{83,84}

The critical time (solid curve) as a function of the peak intensity of the input pump pulse is plotted in Fig.7.2. This curve is analytically obtained by setting the Eqn.(5.2a), $N_e(0, t_{cr}) = N_{cr}$. As one would expect, when the peak intensity is lowered, formation of the overdense plasma layer takes place progressively later along the pulse profile. This dependence is, though pulse-shape dependent, similar to the result from the avalanche model where the breakdown intensity threshold is proportional to reciprocal of the laser pulselength.^{83,84} For pulses with peak intensities $\leq 5 \times 10^{12} \text{ W/cm}^2$, a critical density plasma is formed during the trailing edge of the pulse and a small fluctuation in the intensity (due to fluctuations in the laser pulse energy and pulsewidth) will result in a large change in the critical time. This leads to large fluctuations in probe transmission data as can be seen in Fig.7.3. For higher intensities, by contrast, critical time occurs during the leading edge and a small peak intensity fluctuation leads to only a small change in the critical time, as is the case seen in Figs.4.1 and 4.3 and Figs.5.1 and 5.3.

FIG.7.2: Calculated critical time t_{cr} (solid curve), transmissivity T (dashed curve), and experimentally measured transmissivity (open-dot curve) of the picosecond pump pulse as functions of input peak intensity. Negative (positive) critical time corresponds to formation of the critical density plasma during the leading (trailing) edge of the pulse.

During the time interval ($t_{cr} - t_{sub}$) the laser intensity is sufficiently high to cause ionization of neutral atoms, but still too low to produce a plasma of the critical density at the surface. During the times $t \geq t_{cr}$ when a critical plasma layer is formed at the surface, the subsequently arriving portion of the pulse is completely decoupled from its transmitted portion and is reflected and interacts with the subsequently expanding surface plasma. The transmitted portion traveling at the speed of light and quickly leaving behind the expanding surface plasma suffers no complications due to plasma expansion. For this reason, the transmitted portion rather than reflected portion of the pump pulse is examined to make inference about the ionization rate and temporal narrowing and spectral broadening effects.

FIG.7.3: Frontside probe reflectivity (open dots) and transmissivity (close dots) data taken with the femtosecond laser for uncoated glass targets at average peak intensity $\approx 5 \times 10^{12} \text{ W/cm}^2$. The large statistical fluctuation in the data is due to large fluctuations in the critical time as explained in text.

7.2 Energy and Spectral Measurements and Discussion

Figure 7.2 also shows calculated and measured transmitted pump energies as functions of incident peak intensity. The measured values each are average of 100 laser shots. The calculated values have been averaged over a factor of 4 in intensity to simulate transverse variations in the pump intensity. Transmissivity T of the high intensity pump pulse through transparent targets of 1mm thickness is given by time integrating equations (5.1) and (5.2) and employing the relations $I(\pm\infty)=0$ to obtain the time-integrated solution for the transmitted pump fluence (see Appendix A):

$$\varepsilon(x) = -\frac{1}{a} \ln \left\{ \text{Exp}(-b \int_0^x N_{\infty}(x') dx') [\text{Exp}(-a\varepsilon_{cr}) - 1] + 1 \right\} \quad (7.1)$$

In the above, constants a and b are given in Chapter 5. $\varepsilon(x) = \int_{-\infty}^{+\infty} I(x,t') dt'$ is the transmitted energy fluence, and $\varepsilon_{cr} = \int_{-\infty}^{t_{cr}} I(0,t') dt'$ is the input fluence up to t_{cr} at the surface. As

expected, when the peak intensity is low most of the energy during the pulse is transmitted and little is reflected. Conversely, when the peak intensity is high enough, the overdense plasma layer occurs early during the leading edge, thus limiting transmission of the high intensity laser pulses. This tendency is generally true regardless of the pulshape of pulselength. Figure 7.4 shows the energy measurements of the transmitted pulse taken with the femtosecond laser for different targets and coatings. As was discussed in Chapter 5, because of the absorptive carbon or gold films, the critical plasma layer is formed earlier in the coated target than that in uncoated targets, thus allowing less energy passing through the surface.

FIG.7.4 High intensity transmissivity are measured with the femtosecond laser as functions of peak intensities for different targets.

FIG.7.5: Spectra of transmitted pump pulse are measured at various peak intensities with the femtosecond laser.

The effects of phase modulation and change in the refractive index associated with plasma formation in the transparent medium are further evidenced by observation of spectral shifts and broadening in the transmitted pump pulse. Figure 7.5 displays the spectra of the transmitted portion of the pump pulse at various incident peak intensities. As can be seen, at low peak intensity $I=5 \times 10^{11} \text{ W/cm}^2$ a blip of blue frequency shift of $\sim 3.5 \text{ nm}$ from the peak frequency component is developed. As the intensity is increased, the blue frequency shift is strongly enhanced, further away from the peak component. At the same time, frequency components in the red wings of the spectra are also pronouncedly broadened.

As was discussed in the previous section of this chapter, for intensity $\geq 5 \times 10^{12} \text{ W/cm}^2$ formation of the surface critical plasma always takes place during the leading edge of the laser pulse. As a result, the blue shifts in the spectra with intensity $\geq 5 \times 10^{12} \text{ W/cm}^2$ are solely attributed to a change in the refractive index associated with formation of a plasma in the bulk region; and the red-sided broadening toward solely due to self phase modulation effects associated with the nonlinear refractive index n_2 . According to the Drude's model, the light of wavelength λ will be blue shifted by an amount:

$$\Delta\Phi_p(t) \approx \frac{e^2 L_p \lambda^3}{2\pi m_e c^3} N_e \quad \text{or} \quad \Delta\lambda(t) \approx -\frac{e^2 L_p \lambda^3}{2\pi m_e c^3} \frac{dN_e}{dt} \quad (7.2)$$

where L_p is the length of the bulk plasma, c is the speed of light, and e , m_e and N_e are the charge, mass and plasma density of electrons, respectively. Using $L_p=28 \mu\text{m}$ as the thickness of the bulk plasma of density $>10^{18}/\text{cm}^3$ (see Chapter 5), the corresponding ionization rates can be obtained as a function of peak intensity (Fig.7.6).

FIG.7.6 Measured blue frequency shifts and calculated ionization rate as functions of peak intensity of the incident pump pulse.

FIG.7.7: Measured spectral FWHMs and calculated temporal FWHMs of transmitted portion of the pump pulse as functions of incident peak intensity.

As for the broadening, during the length $L=1\text{mm}$ of the transparent medium with nonlinear refractive factor $\gamma=2.5 \times 10^{-16}(\text{cm}^2/\text{W})$, the phase change $\Delta\Phi$ is accumulated and given by:

$$\Delta\Phi(t) = \frac{2\pi}{\lambda} \gamma \int_0^L I(x,t) dx \quad \text{or} \quad \Delta\lambda(t) = -\frac{2\pi}{\omega} \gamma \int_0^L \frac{dI(x,t)}{dt} dx \quad (7.3)$$

Another contribution to the broadening is attributed to the cut-off in the transmitted pulse as shown in Fig. 7.1. By either contribution or both, the resultant effect is a net shortening in the temporal width of the transmitted pulse which results from spectral broadening. Assuming the transmitted pulse is transform-limited, i.e. $\Delta\nu\Delta\tau=0.5$. The temporal width of the transmitted pulse is estimated as a function of incident peak intensity as shown in Fig.7.7. The results show an initial decrease in the pulsewidth from $\approx 200\text{fs}$ down to a minimum of $\approx 70\text{fs}$ as the intensity increases from 5×10^{10} up to $5 \times 10^{13} \text{W/cm}^2$. At higher intensity, $5 \times 10^{14} \text{W/cm}^2$, the pulsewidth increases back up to $\approx 100\text{fs}$. This behavior can be explained as following: When the peak intensity increases the cut-off in the transmitted pulse occurs sometimes near the half maximum at the leading edge where the intensity change rapidly, effectively resulting in shorter FWHMs of the transmitted pulse. As the intensity is further increased, the cut-off takes place progressively at much earlier time during the leading edge where the intensity profile is rather flat and changes slowly, the resultant FWHM of the transmitted pulse therefore becomes longer.

Clearly the physical picture requires more detailed analysis to account for the spectral shift and broadening due to the competition between the effects of plasma production and self phase modulation. Assuming there is no initial chirping in the laser pulse, the spectral

distribution of the transmitted light of frequency ω_0 can be calculated as, similar to equation (3.3):

$$I(\omega) \sim |\bar{E}(\omega)|^2 = \left| \int_{-\infty}^{+\infty} E(t) e^{i[\Delta\Phi_p(t) + \Delta\Phi(t)]} e^{-i(\omega - \omega_0)t} dt \right|^2 \quad (7.4)$$

As shown in Fig. 7.1 early, prior to the time t_{sub} , laser absorption for plasma production or blue shift is insignificant and the term $\Delta\Phi_p$ is negligible compared to that of SPM. In the time interval $(t_{\text{cr}} - t_{\text{sub}})$ when production of plasma at the surface is rapidly, the plasma effect or the term $\Delta\Phi_p$ is predominant.

In ending this chapter it should be stressed that transmission rather than reflection of the high intensity pump pulses was monitored because of two clear reasons: First, the transmitted pump light passes through the target surface, quickly leaving the surface plasma behind. Thus, it is not complicated by the subsequent expansion of the plasma. Unlike the reflected signals, the transmitted signal is cleaner and free of diffusely scattered light. Another reason is that the transmitted, not the reflected, pump gets absorbed and gives rise to the plasma formation as it travels through the target. Other advantages in monitoring the transmitted pump light can be realized but awaits more reliable high order autocorrelators, e.g. third order autocorrelators, to record the shapes of the cutoff in the transmitted pulse which will provide useful atomic ionization information with high temporal accuracy, less than 20% of the laser pulselength.

CHAPTER 8

CONCLUSION AND PROSPECTS FOR FUTURE WORK

This thesis work has started out with detailed design and construction of experiments for gathering useful information and data about the high temperature and solid state density plasmas produced by ultrashort and high power laser pulse interactions with transparent solid media. A series of different experiments has been carried out so that both time-resolved and time-integrated characterization of the plasma and inference of the important mechanisms may be made. The experimental set-up is still in use and being modified for future experiments to further test the validity of the theoretical models for plasma formation in uncoated and coated transparent solid materials.

The series of experiments performed with intensity below plasma formation threshold has demonstrated the observation of induced phase modulation due to a nonlinear refractive index n_2 in counter-propagating ultrashort pulses. Both experimental and theoretical results show that the induced phase modulation effects can be studied at high intensity (up to $1 \times 10^{13} \text{ W/cm}^2$) by reducing the interaction length to minimize nonlinear effects such as stimulated Raman scattering, self focusing or group velocity reshaping of the pulse.^{60,124} Strongly related to the present study, these experiments have demonstrated a reliable approach to determine the absolute zero timing between the pump and probe pulses within 20% of the pulselength. This thus provides additional information on the timing of the plasma formation processes when laser intensity is increased above the plasma formation threshold.

The frontside probe experiment confirms formation of a super-critical plasma layer at the surface and its subsequent expansion into the surrounding vacuum. The data determine the time scales for the formation (~ 0.5 -1ps) and expansion processes (~ 6 ps). The plasma temperature (~ 40 eV), ionization state ($Z_{av} \sim 4$), and the plasma acoustic velocity of 3×10^6 cm/sec are then inferred from the data using a isothermal plasma expansion. Unfortunately, the frontside probe data shows a serious limitation by the super-critical layer, beyond which the probe light can not penetrate the bulk region and render any additional information on the mechanisms that account for the plasma formation behind the surface.

Complementary to the data gained from frontside probing, experiments with backside probing have been carried out to infer the most important mechanisms in the plasma production. One advantage in backside probing over the frontside probing is that the backside probe light altogether avoids complications associated with plasma expansion such as stimulated Raman and Brillouin scatterings and profile steepening. Another important advantage of the backside probing is that it directly interacts with the bulk region whose changes in optical properties are a manifestation of laser-matter coupling and thermal transport phenomena. Experimental data for uncoated transparent glass demonstrate the laser-matter coupling and formation of a bulk plasma that is caused by inverse Bremsstrahlung absorption and collisional ionization of impurity atoms

The major success of the experiments is the pump and backside probe experiments with targets overcoated with absorptive thin films to eliminate altogether the effects of induced phase modulation and formation of an underdense bulk plasma resulting in further simplification in the analysis. It has demonstrated the feasibility for time-resolved probing of a thermal wave supported by electron conduction. The experimental data, as the first evidence for a supersonically moving (1.8×10^7 cm/s) ionization front in solid

density hot plasma, confirm that electron thermal conduction is the most dominant energy transport mechanism during the first picosecond after the femtosecond laser irradiation. The plasma was found to have a steep gradient and persist for about 15ps, as a manifestation of thermal transport. The physics of such long-lived plasmas, imbedded inside solids can be further explored without complications from hydrodynamic expansion. For example, the plasma-quartz interface in the interior region can be seen as an embodiment of the a Mott-like transition where electrons experience abrupt changes in the material properties, from that of strongly conducting plasma to the insulating quartz. Thermal transport and electrical properties of such interface region remain to be studied. The experimental results demonstrate that a solid hot plasma can be produced, sufficiently tamped and long-lived, such that other investigations should be possible, e.g. solid density plasma spectroscopy, transport properties of strongly coupled and/or Fermi degenerate plasmas, and studies of radiative transfer properties of hot dense matter.

The last experiments consist of the energy and spectral measurements of the high intensity laser pulse transmitted through the transparent targets. These help to determine the timing and intensity of the thresholds for important processes and to characterize the propagation and absorption of such high intensity laser pulses. Also, the spectral measurements give estimates of the ionization rates and importance of the self phase modulation effects due to plasma formation and/or the nonlinearity in the refractive index of the transparent medium.

Future investigations along the lines of this research work require higher power lasers to study in greater detail and with better temporal resolution the effects of the thermal wave propagation in the transparent materials. Also, inhibition of thermal transport in laser-produced plasma has been observed in the past, and the inhibition is attributed to the plasma ablation and other nonlinear processes that are associated with long scale length

plasmas. Studies to isolate and probe the thermal transport should provide more reliable evidence that are not compromised by the plasma expansion. In light of the findings of this research work we propose, as higher intensity lasers now become more available, a series of experiments that are critical for more detailed characterization of the dominant transport mechanism, ionization rates and x-ray emission in laser-produced plasmas in the following sections:

8.1 Electron Thermal Conduction at Higher Intensity Regimes

As was shown in Chapter 6, the pump and backside probe experiment with transparent solid targets advantageously allows direct probing of the bulk region without being complicated by the plasma expansion. Time-resolved frequency shifts that are proportional to the velocity of the electron conduction-supported thermal wave and changes in optical properties of the bulk plasma are recorded with high temporal resolution to infer thermal conductivity and the characteristic time scales for supersonic and subsonic regimes of the thermal wave. The magnitudes of the frequency shifts and reflection enhancements will be increased with the intensity of the absorbed energy delivered to the surface (Chapter 6).

8.2 Localized Measurements of Ionization Rates

As was shown in Chapter 7, the high intensity laser light transmitted through the transparent target suffers a large blue shift and spectral broadening. The blue shift is proportional to the ionization rate in the production of plasma which results in a change in the refractive index of the medium. The spectral broadening toward the red side of the spectrum is due to self phase modulation by the refractive nonlinearity n_2 and is also proportional to the target thickness. Calculations show that with the target thickness in range of $\leq 100\mu\text{m}$ contribution to self phase modulation by n_2 is negligible, and by using

the thickness x variable within this range the ionization rate dN_e/dt is determined from the blue shifts as^{125,126}

$$\Delta\lambda(x) \sim \int_0^x \frac{dN_e(x',t)}{dt} dx' \quad (8.1)$$

Ionization rate per unit atomic layer or localized ionization rate thereby can be obtained by extrapolating the experimental data $\Delta\lambda/\Delta x$, thus making inference of the atomic processes of individual atoms that are exposed to the laser radiation.

8.3 Temporal Mapping of High Intensity Ultrashort Pulses

Along the lines of the experiments described above in section 8.2, temporal profiles of the laser pulses, including any pulse asymmetry, can also be determined from energy measurements of the high intensity light transmitted through the transparent targets through the equality:

$$\frac{dT}{dI_p} = \frac{1}{a} i(t_{cr}) \frac{dt_{cr}(I_p)}{dI_p} \quad (8.2)$$

where a is a proportionality constant, T is the transmissivity measured as a function of incident peak intensity I_p , and t_{cr} is the critical time during the laser pulse when there is a critical density plasma formed at the surface. t_{cr} is a function of I_p and is given by the model discussed in Chapter 5. The normalized intensity profile $i(t)$ is resulted from iterating Eqn.(2) with the experimental data $\Delta T/\Delta I_p$.

Another approach for temporal mapping is to utilize the pump and probe experiment, but with the pump intensity below the plasma formation threshold and still sufficiently high for detectable induced phase modulation in the probe light. The physics, calculations and technical requirements for this approach have been discussed in great details in Chapter 3.

8.4 Risettime of X-Ray Emission

The risetime of x-ray radiation emitted from the laser-produced plasma can be directly measured in the pump and probe experiment described in Sec.8.1, but with some technical modification. The probe light, instead of sensing the plasma region, is now to interact with a transparent region near to the plasma region. The optical properties of the transparent region are not disturbed by the high intensity pump beam, but rather by the x-ray radiation emitted from the nearby plasma. The durations of the measured transient reflection and transmission of the probe light are indicative of the x-ray risetime, though convolved by the finite duration of the probe pulse.

8.5 Localized Measurements of Electron Temperature

In Chapter 6, calculations of electron temperature and density profiles at different depth inside the bulk target are shown by using the nonlinear heat wave model. The penetration depth of the thermal wave is up to some thousands of angstroms. The temperature profiles can be experimentally mapped out by monitoring continuum emission or line signature radiation emitted from a thin layer of well known atomic elements. The thin layer is buried inside the target with a variable distance from the target surface to provide space-resolved detection of the emitted radiation. Also, the electron density profiles can also be deduced from Stark broadening of the line emission.^{122,127} These measurements first will whether further confirm the validity of the nonlinear heat wave model proposed in Chapter 6 or the nonlocality of the electron thermal transport in a high temperature and solid density plasma. And finally, the measurements without complication by plasma expansion, will allow to determine whether the electron thermal transport is strongly inhibited as was reported in the past with so much confusion over the specific mechanisms associated with plasma expansion. If there is any significant inhibition, the model will help to characterize the degree of inhibition.

APPENDIX A

In the first section of this appendix I show algebraic details of an analytical approach to the two equations in Chapter 5 for plasma formation in uncoated transparent solid materials. The model is a modification of the well-known avalanche model. The model gives both time-dependent and time-integrated solutions to the electron density and the transmitting high intensity laser (pump) pulse in the bulk material. And at the same time, it preserves characteristics of the avalanche model. In section A2 derivation of an approximate analytical solution to the nonlinear heat equation in Chapter 6 for electron temperature profile is given. The solution is self-similar due to the highly nonlinear temperature dependence of the electron thermal conductivity. Quantities are expressed in cgs units unless specified otherwise.

A1: The modified avalanche model

Integrating Eqn.(5.2), without the term in $\partial T_e / \partial t$ and the terms in the bracket, we obtain the density as a function of both space and time as

$$N_e(x, t) = N_{e0}(x) e^{-a \int_{-\infty}^t I(x, t') dt'} \quad (A1)$$

where $N_{e0} = N_e(x, -\infty)$ is the initial electron density distribution. When the target is initially homogeneously uniform, $N_{e0} = \text{constant}$. Putting this into the right hand side of Eqn.(5.1), integrating from $-\infty$ and employing the condition $I(x, -\infty) = 0$ we obtain:

$$I(x,t) + \frac{c}{n} \frac{\partial}{\partial x} \int_{-\infty}^t I(x,t') dt' = \frac{bc}{n} N_{eo}(x) \int_{-\infty}^t I(x,t') e^{-a \int_{-\infty}^t I(x,t'') dt''} dt' \quad (A2)$$

Let $\xi(x,t) = \int_{-\infty}^t I(x,t') dt'$ then $\frac{d\xi}{dt} = I(x,t)$ and $\xi(x, -\infty) = 0$

The right hand side of (A2) can be rewritten as:

$$\text{RHS(A2)} = -\frac{bc}{n} N_{eo}(x) \int_{-\infty}^{\xi(x,t)} e^{a\xi'} d\xi' = \frac{bc}{na} N_{eo}(x) \left(1 - e^{-a\xi(x,t)}\right)$$

$$\text{Thus } \frac{\partial \xi}{\partial t} + \frac{c}{n} \frac{\partial \xi}{\partial x} = \frac{bc}{na} N_{eo}(x) \left(1 - e^{-a\xi(x,t)}\right) \quad (A3)$$

Along the characteristic curve $x=c/n(t-t_0)$ and $dx/dt=c/n$, (A3) can be rewritten as:

$$\frac{d\xi}{dt} = \frac{bc}{na} N_{eo}(x(t)) \left(1 - e^{-a\xi(x(t),t)}\right)$$

This describes the energy transmission of the laser (pump) pulse, whose motion follows the characteristic line, i.e. at $x(t_0) = 0$ the pump pulse begin its interaction with the medium. Rewrite this as:

$$\frac{d}{dt}(e^{-a\xi}) + \frac{bc}{n} N_{eo} e^{-a\xi} = \frac{bc}{n} N_{eo}$$

$$\text{or } \frac{d}{dt} \left(e^{-a\xi} e^{\frac{bc}{n} \int_{t_0}^t N_{eo}(x(t')) dt'} \right) = \frac{bc}{n} N_{eo} e^{\frac{bc}{n} \int_{t_0}^t N_{eo}(x(t')) dt'}$$

Integrating from t_0 to t along the characteristic line, $t_0 = t - nx/c$

$$e^{-a\xi} e^{\frac{bc}{n} \int_{t_0}^t N_{eo}(x(t')) dt'} - e^{-a\xi(0, t - nx/c)} = \frac{bc}{n} \int_{t_0}^t N_{eo}(x(t')) e^{\frac{bc}{n} \int_{t_0}^t N_{eo}(x(t')) dt'} dt'$$

or

$$e^{-a\xi} e^{b \int_0^x N_{eo}(x') dx'} - e^{-a\xi(0, t - nx/c)} = b \int_0^x N_{eo}(x') e^{b \int_0^x N_{eo}(x') dx'} dx'$$

$$\text{Let } \eta(x) = b \int_0^x N_{eo}(x') dx' \quad \text{and} \quad d\eta = b N_{eo}(x) dx$$

$$e^{-a\xi} e^{\eta} - e^{-a\xi(0, t - nx/c)} = e^{\eta} - 1 \quad (\text{A4})$$

From (A1) we obtain

$$N_e(x, t) = \frac{N_{eo}(x)}{1 + e^{-\eta} (e^{-a\xi(0, t - nx/c)} - 1)} \quad (\text{A5})$$

and from (A4)

$$\xi(x, t) = -\frac{1}{a} \ln \left[1 + e^{-a\xi} \left(e^{-a\xi(0, t - nx/c)} - 1 \right) \right] \quad (\text{A6})$$

$$I(x, t) = \frac{\partial \xi}{\partial t} = \frac{I_o(t - nx/c)}{1 - e^{-a\xi(0, t - nx/c)} (1 - e^{\eta(x)})} \quad (\text{A7})$$

Rewrite (A5) and (A7) for time-dependent solution for the electron density and intensity of the high intensity pump pulse up to the critical time t_{cr} . t_{cr} is defined as the time when a critical plasma is formed at the surface, i.e. $N_e(0, t_{cr}) = N_{cr}$:

$$N_e(x, t) = \frac{N_{e0}(x)}{1 + e^{-b \int_0^x N_{e0}(x') dx' - a \int_{-\infty}^{t - nx/c} I_0(t') dt' - 1}} \quad (A8)$$

and

$$I(x, t) = \frac{I_0(t - nx/c)}{1 - e^{-a \int_{-\infty}^{t - nx/c} I_0(t') dt' - b \int_0^x N_{e0}(x') dx' - 1}} \quad (A9)$$

The time-integrated solution for the total energy fluence transmitted through a medium of thickness x is given by (A6):

$$\xi(x) = -\frac{1}{a} \ln \left[1 + e^{-b \int_0^x N_{e0}(x') dx' \left(e^{-a \xi_{cr}} - 1 \right)} \right] \quad (A10)$$

Where $\xi(x, t) = \int_{-\infty}^{t_{cr}} I_0(0, t') dt'$ is the input fluence up to t_{cr} at the surface

Because of the integrals in (A8) and (A9), it often assumes integratable function Sech^2 as the laser pulseshape. Note that this is merely a computational convenience because there is little or essentially no significant difference between Gaussian or Sech^2 functions as shown in Chapter 2. Another feature of the model is that the transmitted fluence is independent of the pulseshape for the laser pulse as shown in (A10).

A2: Approximate analytical solution to the nonlinear heat equation

Following the lines of Marshak²⁹ we solve the total internal energy described by Eqn.(6.4) subjected to the boundary conditions $\varepsilon(0,t) = \varepsilon_0 \text{Exp}(2\gamma t)$ and $\varepsilon(x, -\infty) = 0$.

During the laser heating, $t \leq 0$, by using a similar form for the solution:

$$\varepsilon(x,t) = \varepsilon_0 \text{Exp}(2\gamma t) f(z) \quad \text{where } f(z) = x \text{Exp}(-q\gamma t) \quad \text{for } t \leq 0 \quad (\text{A11})$$

and using the operators:

$$\frac{\partial}{\partial x} = \frac{\partial z}{\partial x} \frac{d}{dz} = \text{Exp}(-q\gamma t) \frac{d}{dz} \quad \text{and} \quad \frac{\partial}{\partial t} = \frac{\partial z}{\partial t} \frac{d}{dz} = -q\gamma z \frac{d}{dz}$$

Eqn.(6.4) is transformed into a nonlinear ordinary differential equation:

$$qz \frac{df(z)}{dz} + \frac{c_1 \varepsilon_0^q}{\gamma} \frac{d^2}{dz^2} f^{q+1}(z) - 2f(z) = 0 \quad (\text{A12})$$

subjected to new boundary conditions : $f(0) = 1$, and $f(\infty) = 0$. Note that for $q=0$ (A12) corresponds to an ordinary heat transfer equation. Also since the first term is proportional to z , the solution does not exist as $z \rightarrow \infty$. Therefore, $f(z)$ must have a finite extension, i.e. $f(z) = 0$ for $z > z_f$ where z_f is determined from the energy conservation Eqn.(6.5). Now consider solution to (A12) with $q > 0$ and expand f in a Taylor series about z_f , i.e.

$$f(z) = a \left(1 - \frac{z}{z_f}\right)^{m_1} \left[1 + b \left(1 - \frac{z}{z_f}\right)^{m_2} + \dots\right] \quad (\text{A13})$$

where a and b are constants, given by the boundary conditions. Substituting this into (A12), selecting terms of same power in f , and neglecting the second term in (A13) we obtain the solution to z_f and f , or solutions to ε and x_f as shown in Chapter 6.

For $t > 0$ or after the laser heating, the heated thin surface layer is treated as an instantaneous source for subsequent diffusion of heat further into the colder region. In this view Eqn.(6.4) with the boundary conditions

$$\frac{\partial}{\partial x} \varepsilon(0, t) = 0 \quad \text{and} \quad \varepsilon(+\infty, t) = 0 \quad (\text{A14})$$

can be solved by using a similar transformation :

$$z = \frac{x}{(k_c F_a^q t)^{1/(2+q)}} \quad \text{and} \quad \varepsilon(x, t) = \left(\frac{F_a^2}{k_c t} \right)^{1/(2+q)} f(z) \quad (\text{A15})$$

in the same way described above for $t \leq 0$, substituting these into (6.4) to obtain an ordinary nonlinear differential equation for f , i.e.

$$(q+2) \frac{d}{dz} \left(f^q \frac{df}{dz} \right) + z \frac{df}{dz} + f(z) = 0 \quad (\text{A16})$$

Trying the form $f(z) = [A(z_f^2 - z^2)]^{1/q}$ to (A16) using boundary conditions described above and the energy balance Eqn.(6.5) to obtain solutions to f and z_f or solutions to ε and x_f after laser heating as shown in Chapter 6

APPENDIX B

The following program was written in FORTRAN Language to run on VAX computer mainframe at LLNL and utilizes the available math software package IMSL to perform Fast Fourier Transform. The numerical output is written on an output file for later graphic display. The program calculates frequency shifts due to induced phase modulation during either or both counter- and co-propagating journeys, as discussed in Chapter 3, on the probe that sees a time-dependent refractive index induced by the strong pump pulse. The calculation is performed at each time delay and can be used either a Gaussian or Sech² pulseshape. Also the program calculates the autocorrelation function of the probe pulse as given by Eqn.(2.1), Chapter 2. All calculations are performed with double precision digits.

```

PROGRAM IPM_CAL
IMPLICIT REAL*8 (A-H,O-Z)
PARAMETER(NEXP=12,NWSAMP=2**NEXP,NTDLY=50,NSTART=1900,
NLAST=NWSAMP-NSTART+1)
COMPLEX*16 FUN(NWSAMP)
DIMENSION TD(NTDLY),DF(NTDLY),F(NWSAMP),EE(NWSAMP),
      TEMP1(NWSAMP),TEMP2(NWSAMP),IWK(NEXP+1),AUTO(NTDLY)
COMMON
/INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,PULSEWIDTH,FAC,RATIO,
      A,B
COMMON /GAUSS/EP(5),WG(5)
OPEN(UNIT=11,FILE='{VU.XPM}XPM1_DW.DAT',STATUS='NEW')
CALL GETINPUT
DO 100,I=1,NTDLY
      TD(I)=T0+DBLE(I-1)*DTDLY
100  CONTINUE
DO 110 I=0,NWSAMP-1
      F(I+1)=DBLE(I)/TPERIOD-NWSAMP/(2.*TPERIOD)
110  CONTINUE
DO 200, I=1,NTDLY      !AT EACH TIME DELAY
      CALL GETFUN(TD(I),NWSAMP,FUN)
      CALL FFT2C(FUN,NEXP,IWK)
      CALL FINDMAX_IW(TD(I),NWSAMP,F,FUN,TEMP2,EE,DF(I))
      CALL
      REARRANGE(NWSAMP,NSTART,NLAST,F,EE,NUM,TEMP1,TEMP2)
      CALL AUTOCORR(TD(I),AUTO(I))
      WRITE(11,201)TD(I),DF(I),AUTO(I)

```

```

WRITE(11,202)TD(I),DF(I)
202 FORMAT(1X,F5.2,2X,F8.5)
200 CONTINUE      !NEXT TIMEDELAY
RISET=PULSEWIDTH/2.
FALLT=RISET/DSQRT(FAC)
WRITE(11,203)ALFA,BETA,BETAOFF,RATIO,RISET,FALLT
203FORMAT(1X,'ALF',F8.4,'BET',F8.4,'BETAOFF',F5.2,/, 'L1/L',F5.2,'RISET',
F5.2,'FALLT',F5.2) !L1:INT LENGTH FOR INCOMING JOURNE
WRITE(11,*)
END      ! *****
SUBROUTINE GETINPUT
IMPLICIT REAL*8 (A-H,O-Z)
COMMON /INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,
PULSEWIDTH,FAC,RATIO,A,B
COMMON /GAUSS/EP(5),WG(5)
EP(1)=.1488743390
EP(2)=.4333953941
EP(3)=.6794095683
EP(4)=.8650633667
EP(5)=.9739065285
WG(1)=.2955242247
WG(2)=.2692667193
WG(3)=.2190863625
WG(4)=.1494513492
WG(5)=.0666713443
T0=-2.
DTDLY=.1
TPERIOD=100.
RATIO=0.6      !1(0)TO TURN ON(OFF) RETURN JOURNE
ALFA=.45      !RATIO FOR INCOMING/RETURN,IF =0:NO INCOMING JOURNEY
BETA=2.6
BETAOFF=0.      !=0. NO RETURN CONTRIBUTION
PULSEWIDTH=0.44
A=2.*DLOG(DBLE(2.))/PULSEWIDTH/PULSEWIDTH
B=0.
FAC=1./4.      !FAC>1 (OR <1 ) , PULSEWIDTH DECREASES (OR INCREASES)
RETURN
END      ! *****
DOUBLE COMPLEX FUNCTION EFIELD(T)
IMPLICIT REAL*8 (A-H,O-Z)
COMPLEX*16 CI
PARAMETER (CI=(0.,1.))
COMMON /INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,
PULSEWIDTH,FAC,RATIO,A,B
IF (T.GT.0.) THEN
EFIELD=CDEXP(-FAC*(A+B*CI)*T*T)
ELSE
EFIELD=CDEXP(-(A+B*CI)*T*T)
ENDIF
RETURN
END      ! *****
SUBROUTINE GETFUN(TDLY,NWSAMP,FUN)
IMPLICIT REAL*8 (A-H,O-Z)
COMPLEX*16 FUN(NWSAMP),EPUMP,CI,EFIELD,FASE

```

```

PARAMETER (CI=(0.,1.))
COMMON /INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,
      PULSEWIDTH,FAC,RATIO,A,B
DT=TPERIOD/NWSAMP
DO 1000 J=0,NWSAMP-1
TIME=DBLE(J)*DT-TPERIOD/2.
TTD=TIME+TDLY
EPUMP=EFIELD(TTD)
IF (ALFA.GT.0.) THEN
TTD0=TTD-RATIO/ALFA
CALL GAUSINT(0.,TTD,FASE1)
CALL GAUSINT(TTD0,0.,FASE0)
FASE=BETA*ALFA*(FASE1+FASE0)
ELSE
FASE=0.
ENDIF
IF(BETAOFF.NE.0.) FASE=FASE+BETA*(1.-RATIO)*EPUMP*DCONJG(EPUMP)
FUN(J+1)=EFIELD(TIME)*CDEXP(CI*FASE)
1000 CONTINUE
RETURN
END      ! *****
SUBROUTINE PLOT3D(TDLY,Y,Z,NUM)
IMPLICIT REAL*4 (A-H,O-Z)
REAL*8 Y(600),Z(600),TDLY
DIMENSION YS(600),ZS(600),TS(600)
DO 6000,K=1,NUM
TS(K)=SNGL(TDLY)
YS(K)=SNGL(Y(K))
ZS(K)=SNGL(Z(K))
6000 CONTINUE
RETURN
END      ! *****
SUBROUTINE FINDMAX_IW(TDELAY,NWSAMP,F,FUN,TEMP,EE,FREQSHIFT)
IMPLICIT REAL*8 (A-H,O-Z)
REAL*8 F(NWSAMP),EE(NWSAMP),TEMP(NWSAMP)
COMPLEX*16 FUN(NWSAMP)
COMMON /INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,
      PULSEWIDTH,FAC,RATIO,A,B
DO 3000,K=1,NWSAMP
TEMP(K)=FUN(K)*DCONJG(FUN(K))*(TPERIOD/NWSAMP)**2
3000 CONTINUE
CREARRANGE FOURIER TRNAS ARRAY:
DO 3010 I=1,NWSAMP/2
3010 EE(I)=TEMP(NWSAMP/2+I)
DO 3020 I=1,NWSAMP/2
3020 EE(I+NWSAMP/2)=TEMP(I)
PEAKI=0.
DO 3030 K=1,NWSAMP
IF(EE(K).GT.PEAKI) THEN
PEAKI=EE(K)
INDEX=K
ENDIF
3030 CONTINUE
FREQSHIFT=F(INDEX)

```



```

CWRITE(11,*)'TDELAY=',TDELAY,F=',F(INDEX)
WRITE(6,*)'TDELAY=',TDELAY,F=',F(INDEX)
RETURN
END      ! *****
SUBROUTINE
REARRANGE(NWSAMP,NSTART,NLAST,F,EE,NUM,TEMP1,TEMP2)
IMPLICIT REAL*8 (A-H,O-Z)
DIMENSION TEMP1(NWSAMP),TEMP2(NWSAMP),F(NWSAMP),EE(NWSAMP)
NUM=NLAST-NSTART+1
DO 4000 L=1,NUM
TEMP1(L)=F(NSTART+L-1)
4000 TEMP2(L)=EE(NSTART+L-1)
RETURN
END      ! *****
SUBROUTINE GAUSINT(A,B,VAL)
IMPLICIT REAL*8 (A-H,O-Z)
COMPLEX*16 EFIELD
COMMON /GAUSS/EP(5),WG(5)
C=(B+A)/2.
D=(B-A)/2.
VAL=0.
DO 3000, L=1,5
X1=C+D*EP(L)
X2=C-D*EP(L)
VAL=VAL+WG(L)*(DREAL(EFIELD(X1)*DCONJG(EFIELD(X1)))+
DREAL(EFIELD(X2)*DCONJG(EFIELD(X2))))
3000 CONTINUE
VAL=VAL*D
RETURN
END      ! *****
SUBROUTINE AUTOCORR(TDLY,VAL)
IMPLICIT REAL*8 (A-H,O-Z)
COMPLEX*16 EFIELD
COMMON /INPUT/T0,DTDLY,TPERIOD,ALFA,BETA,BETAOFF,
PULSEWIDTH,FAC,RATIO,A,B
T12=TPERIOD/2.
NPT=10000
DT=TPERIOD/NPT
VAL=0.
DO 5000, L=2,NPT
T=-T12+DBLE(L-1)*DT
TTD=T+TDLY
VAL=VAL+DT*DREAL(EFIELD(T)*DCONJG(EFIELD(T)))*DREAL(EFIELD(TTD)*DCONJG(EFIELD(TTD)))
5000 CONTINUE
VAL=VAL*2.
TTD=-T12+TDLY
VAL=VAL+DT*DREAL(EFIELD(-T12)*DCONJG(EFIELD(-T12)))*
DREAL(EFIELD(TTD)*DCONJG(EFIELD(TTD)))
TTD=T12+TDLY
VAL=VAL+DT*DREAL(EFIELD(T12)*DCONJG(EFIELD(T12)))*
DREAL(EFIELD(TTD)*DCONJG(EFIELD(TTD)))
RETURN
END

```

APPENDIX C

The following program was originally written in Basic Language to run on a IBM computer by W. Cook (LLNL) in 1986, then rewritten by O. L. Landen and later was modified by B.-T. V. Vu to adjust to the needs and conditions of this work. The program and the IBM are used to raster the target at the rate the laser systems fire 10 laser shots per second. The program is to introduce variable parameters to translate the time delay line within a given interval. At the same time it sends commands to raster the target on a travel of 50mm. Velocity and distance are in mm/s and mm, respectively. All raster drivers are Newport products, used with translator mechanisms and target holder that I designed. The fast horizontal actuator, model 850AV-2, is customized with actual velocity equal to 6.4 times the velocity reading and maximum velocity equal to 1.92mm/sec. The actuators used for vertical raster and delay line, model 850A-2, are standard ones with maximum velocity equal to 0.4mm/sec. The actuator driver is a programmable controller, model 855C.

```

10 KEY OFF:COLOR 15,2,1:CLS
11 '
15 ON ERROR GOTO 500
20 DIM POSITION$(6),POSITION(6)
30 LOCATE 10,14:PRINT "WELCOME TO THE NRC 855C MOTORMIKE
CONTROLLER PROGRAM"
35 '
40 OPEN "COM1:600,N,8,,CS,DS,CD" AS #1
45 '
50 C$="!"
55 GOSUB 4000
60 GOSUB 7100 'DELAY LOOP
65 GOSUB 6000
70 IF P$="" THEN 50
71 A$=".4"
72 PRINT #1,A$
73 CLS
75 LOCATE 14,10
76 INPUT "TYPE A FOR AUTO (A) ";Q$
701 ' ***THIS IS THE AUTOMATIC MODE RASTER SETUP ROUTINE***
703 '

```

```

705 MARKER=2
706 CLS:LOCATE 10,22
707 INPUT "RASTER START POSITION (XX.XXXX MM)";INC$
708 INC=VAL(INC$)
709 '
710 ' *** CONVERT MM TO MICRONS ***
711 '
712 INC=INC*10000
713 '
714 PRINT #1,"V10.3"
715 PRINT #1,"V20.2"
716 PRINT #1,"M10.0"
717 PRINT #1,"M2" + INC$
719 GOSUB 2000      'PRINT STATIC AXES POSITIONS
720 GOSUB 1000      'READ CURRENT POSITIONS OF AXES FROM KEYPAD
721 GOSUB 3000      'PRINT CURRENT AXIS POS.
722 M1=VAL(POSITION$(1))*10000
723 M2=VAL(POSITION$(2))*10000
724 '
725 ' *** CHECK IF MOTORS ARE AT DESTINATION ***
726 '
727 IF M1>-10 AND M1<10 AND M2>INC-5 AND M2<INC+5 THEN 728 ELSE 720
728 '
729 CLS:LOCATE 10,22
730 INPUT "TIMEDELAY START POSITION (XX.XXXX MM)";S$
731 S=VAL(S$)
732 GOSUB 5100
733 LOCATE 12,22
734 INPUT "TIMEDELAY END POSITION (XX.XXXX MM)";E$
735 E=VAL(E$)
736 GOSUB 5200
737 LOCATE 14,22
738 INPUT "VERTICAL AXIS INCREMENT (XX.XXXX MM)";INC1$
739 INC1=VAL(INC1$)
740 INC1=INC1*10000
741 '
742 ' *** CALCULATE TIMEDELAY LINE VELO. ***
743 '
744 D=E-S
745 V=D/26.8: IF V>.4 THEN 746 ELSE 747
746 PRINT "SORRYTIMEDELAYRANGE IS TOO LARGE.REENTER IT":GOTO 729
747 V=INT(V*10000)
748 ' *** CONVERT TO STRING VARIABLE WITH FIRST CHARACTER
REMOVED ***
749 '
750 V$=STR$(V)
751 L=LEN(V$)
752 L=L-1
753 V$=RIGHT$(V$,L)
754 '
755 S=S*10000:E=E*10000
756 S$=STR$(S):E$=STR$(E)
757 L=LEN(S$):L=L-1:S$=RIGHT$(S$,L)
758 L=LEN(E$):L=L-1:E$=RIGHT$(E$,L)

```

```

759 '
760 ' *** SEND DELAYLINE TO START POSITION ***
761 '
762 PRINT #1,"V30.4"
763 PRINT #1,"M3" + S$
764 '
765 CLS
766 GOSUB 2000      'PRINT STATIC SCREEN
767 GOSUB 1000      'POLLING CURRENT POS FROM CONTROLLER
768 GOSUB 3000      'AND PRINT
769 '
770 ' *** CHECK IF DELAY LINE IS AT DESTINATION ***
771 '
772 M=VAL(POSITION$(3))
773 M=M*10000
774 IF M>S-10 AND M<S+10 THEN 775 ELSE 767
775 '
776 GOSUB 3000
777 PRINT #1,"V3" + V$
778 '
779 LOCATE 20,26:PRINT "PRESS <G> TO START SEQUENCE."
781 LOCATE 22,31:PRINT "PRESS <X> TO EXIT."
790 K$=INKEY$
791 IF (K$="G" OR K$="g") AND MARKER=1 THEN 920
792 IF (K$="G" OR K$="g") AND MARKER=2 THEN 910
793 IF K$="X" THEN 600 ELSE 790
795 '
796 ' *** PRINT ERROR MESSAGE ***
797 '
798 LOCATE 18,21
799 PRINT "SORRY-THAT POSITION IS OUT OF RANGE-TRY AGAIN"
800 GOSUB 7200
801 GOTO 729
802 '
805 '      ***THIS IS THE RASTER DRIVER ROUTINE***
807 '
910 PRINT #1,"M17.8431"      'TRAVEL 50MM
911 PRINT #1,"M3" + E$
913 MARKER=1
914 GOTO 960
915 '
920 PRINT #1,"M10.0"
921 PRINT #1,"M3" + E$
923 MARKER=2
924 GOTO 960
925 '
926 ' *** QUERY TO RUN AGAIN ***
927 '
930 GOSUB 1000:GOSUB 2000:GOSUB 3000
940 LOCATE 16,27
941 INPUT "DO YOU WANT TO GO AGAIN";Q$
942 IF Q$="N" THEN 600 ELSE 945
945 LOCATE 18,23
946 INPUT "DO YOU WANT TO CHANGE THE SETUP";Q$

```

```

947 IF Q$="Y" THEN 729 ELSE 775
960 '
961 ' ***.UPDATE SCREEN ***
962 '
963 GOSUB 1000
964 GOSUB 3000
965 M$=POSITION$(3)
966 M=VAL(M$)
967 M=M*10000
968 IF M>E-10 AND M<E+10 THEN 980 ELSE 960
969 '
970 ' *** SEND DETECTOR BACK TO START POSITION ***
971 '
980 PRINT #1,"V30.4"
981 PRINT #1,"M3"+S$
985 GOSUB 1000
986 GOSUB 3000
987 M$=POSITION$(3)
988 M=VAL(M$)
989 M=M*10000
990 IF M<S+10 AND M>S-10 THEN 994 ELSE 985
991 '
992 ' *** INCREMENT VERTICAL POSITION ***
993 '
994 INC=INC + INC1
995 INC$=STR$(INC)
996 L=LEN(INC$)
997 L=L-1:INC$=RIGHT$(INC$,L)
998 PRINT #1,"M2" + INC$
999 GOTO 930
1001 '
1003 ' ***THIS IS THE POSITION POLLING SUBROUTINE***
1005 '
1100 FOR Y=1 TO 4
1200 C$="C" + CHR$(Y+48)
1201 AXIS=Y
1300 GOSUB 4100
1400 N=LOC(1)
1500 IF N<10 THEN 1400
1600 GOSUB 6000
1601 POSITION$(AXIS)=P$
1700 NEXT
1800 RETURN
2000 '
2003 ' ***THIS IS THE STATIC SCREEN PRINT ROUTINE***
2006 '
2100 CLS
2101 LOCATE 6,30
2102 PRINT "AXIS #1"
2103 LOCATE 8,30
2104 PRINT "AXIS #2"
2105 LOCATE 10,30
2106 PRINT "AXIS #3":LOCATE 10,52 : PRINT"= TIMEDELY ="
2107 LOCATE 12,30

```

```

2108 PRINT "AXIS #4"
2109 RETURN
3003 ' - ***THIS IS THE DYNAMIC SCREEN PRINT SUBROUTINE***
3100 LOCATE 6,43
3101 K=VAL(POSITION$(1)):K=K*6.375:PRINT USING " ##.####";K
3102 LOCATE 8,43
3103 J=VAL(POSITION$(2)):PRINT USING " ##.####";J
3104 LOCATE 10,43
3105 PRINT POSITION$(3):T=VAL(POSITION$(3)):T=T/.15
3106 LOCATE 10,64:PRINT USING " ##.####";T: LOCATE 10,73:PRINT "ps"
3107 LOCATE 12,43: J=VAL(POSITION$(4)):J=-J*2
3108 PRINT USING " ##.####";J
3109 RETURN
4001 '
4003 '      ***THIS IS THE COMMAND OUTPUT SUBROUTINE***
4100 PRINT #1,C$
4101 RETURN
5001 '
5003 '      ***THIS IS THE SOFTWARE LIMIT CHECKING SUBROUTINE***
5100 IF S<=50 AND S>=0 THEN RETURN ELSE GOTO 795
5200 IF E<=50 AND E>=0 THEN RETURN ELSE GOTO 795
5250 IF F<70.1 AND F>0.0 THEN RETURN ELSE GOTO 7500
5300 IF AXIS=1 AND D<=50 THEN RETURN ELSE 5400
5400 IF AXIS=2 AND D<=45 THEN RETURN ELSE 5500
5500 IF AXIS=3 AND D<=48 THEN RETURN ELSE 5600
5600 LOCATE 20,18
5601 PRINT "SORRY-THAT POSITION IS OUT OF RANGE-TRY AGAIN"
5602 GOSUB 7200
5603 LOCATE 20,18
5604 PRINT "
5605 LOCATE 18,46
5606 PRINT "
5607 GOTO 440
6001 '
6003 '      ***THIS IS THE RESPONSE INPUT SUBROUTINE***
6006 '
6100 L=LOC(1)
6101 IF L=0 THEN RETURN
6200 P$=INPUT$(LOC(1),#1)
6201 RETURN
7001 '
7003 '      *** DELAY LOGPS***
7100 FOR DELAY=0 TO 120
7101 NEXT
7102 RETURN
7103 '
7200 FOR DELAY=0 TO 1000
7201 NEXT
7202 RETURN

```

APPENDIX D

The following is a geometrical analysis for horizontal and vertical alignment of the target, so that as the target is rastered throughout a 5cmx5cm scan, overlap between the pump and probe beams are maintained. This is to ensure the same condition of plasma formation and probing on each laser shot. By iterating the alignment procedure walkoff between the two beams after a scan can be kept below 20 μ m, much smaller than the crater diameter of 75 μ m.

Horizontal alignment process begins with two distant craters c1 and c2 made on a horizontal line at the surface and two He-Ne laser beam as shown in Fig. D1.

FIG.D1: Top view of the target holder for horizontal alignment

Notations are defined as :

- a: the angle between the tilted target relative to the horizontal raster direction.
- b: angle between the two He-Ne laser beams.
- T_0 : initial position of the target when two beams are focused at the crater c1 & c2.

- T₁, T₂: positions of the target after first and second moves.
- d₁: the distance to move the target to position T₁, beam B₁ & the crater c₂.
- d₂: the distance to move the target to position T₂, beam B₂ & the crater c₂.

The beam B₁ is aligned and made to be normally incident at the surface. From the beam-target geometry we determine the tilt angle α :

$$\tan(\alpha) = \frac{d_2 - d_1}{d_1 \tan(b)} \quad (D.1)$$

The tilt angle is then used to readjust the target parallel to the direction of the horizontal raster. This is done by the horizontal tilt micrometer shown in the actual design of the target holder (Chapter 2).

Vertical alignment: The geometrical analysis is the same as the horizontal alignment but with two craters made on a vertical line for calculating the tilt angle between the target and the vertical raster direction.

APPENDIX E

The following are formulae often used to obtain estimates of important plasma and laser parameters in the experiments. All quantities are in cgs unit except temperature expressed in eV. Laser electric field and intensity are in V/cm and W/cm², respectively. A is ion mass in units of proton mass or atomic number. γ is the specific heat per unit volume.

Number density of atoms in SiO₂ solid : $n_a = 6.1 \times 10^{22}/\text{cm}^3$

Critical density : $n_{cr} = 3.14 \times 10^{-10} \omega^2$; for $\omega = 3.1 \times 10^{15} \text{Hz}$; $n_{cr} = 3.02 \times 10^{21} / \text{cm}^3$

$$\text{Electron plasma frequency : } \omega_{pe} = 5.64 \times 10^4 \sqrt{n_e} \quad (\text{E1})$$

$$\text{Plasma skin depth : } \delta = \frac{c}{2\omega_{pe}} = \frac{5.31 \times 10^5}{\sqrt{n_e}} \quad (\text{E2})$$

$$\text{Acoustic (ion sound) velocity : } c_s = 9.5 \times 10^5 \sqrt{\frac{\gamma Z T_e}{A}} \quad (\text{E3})$$

$$\text{Thermal electron velocity : } v_e = \sqrt{\frac{1.5 k T_e}{m_e}} = 4.19 \times 10^7 \sqrt{T_e} \quad (\text{E4})$$

$$\text{Electron-electron collision time}^{13}: \tau_{e-e} = 3.3 \times 10^4 \frac{T_e^{1.5}}{n_e} \quad (\text{E5})$$

Electron-ion energy transfer time¹³: $\tau_{e-i} = 3.5 \times 10^7 A \frac{T_e^{1.5}}{Z n_e}$ (E6)

3-body recombination time⁸¹: $\tau_3 = 1.8 \times 10^{26} A \frac{T_e^{4.5}}{n_e^2}$ (E7)

Gaussian laser pulse shape: $I(t) = I_0 e^{-\alpha t^2}$; with FWHM $\Delta\tau = \sqrt{\frac{4 \ln 2}{\alpha}}$ (E8)

Sech² laser pulse shape : $I(t) = I_0 \text{Sech}^2(\alpha t)$; with FWHM $\Delta\tau = 1.76275/\alpha$ (E9)

Laser electric field : $E \approx 27\sqrt{I}$ (E10)

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