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Interaction Studies with Intense,
Ultra-short Pulses

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Production of dense vapor targets for laser-plasma interaction studies with intense, ultra-short pulses

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ABSTRACT

The technique of laser-induced ablation of thin films from glass slide substrates has been investigated as a candidate vapor target production method for studies of both tunneling-driven x-ray/xuv recombination lasers and relativistic propagation using intense, ultra-short laser pulses. It is shown by simultaneous two-wavelength interferometry that particle densities of order $10^{19}/\text{cm}^3$ are readily achieved and that some intrinsic ionization accompanies the plume formation. Absorption measurements with both 100 picosecond and 125 femtosecond pulses are consistent with observed edge velocities near 10^6 cm/sec. The level of ionization driven by the intense 125 femtosecond laser pulse has been coarsely estimated. Averaged estimates from spectral blue shifting of spectra transmitted through the plume are consistently lower than those obtained from evaluation of saturation intensity thresholds based on the sequential nonresonant optical field ionization (OFI) process.

2. INTRODUCTION

The primary goal of our work is the study of tunneling-driven (i.e. strong optical field ionization), recombination x-ray / xuv lasers and associated relativistic propagation phenomena at high peak laser intensities in the ultra-short pulsewidth (near 125 femtoseconds) regime. This requires reliable production of vapor targets typically in the subcritical density range, 10^{19} to 10^{21} cm^{-3} . Furthermore, any ionization related to the recombination x-ray laser problem is to be characterized by relatively cool photoelectron temperatures (typically of order 10's of electron volts) to enhance the three body recombination process at high density. These two requirements indicate that such plasma production (allowing for potential intrinsic ionization in vapor formation) approaches the strongly coupled regime where Γ , the ratio of Coulomb binding to thermal energy exceeds unity. That is,

$$\Gamma = \frac{Ze^2}{KT} \left(\frac{4\pi n_e}{3} \right)^{\frac{1}{3}} \geq 1 \quad (1)$$

where Ze is the ion charge state, KT is the thermal energy, and n_e is the electron density.

A variety of gas nozzle designs exist for experiments with noble gas jets at high density. As a complement to the gas nozzle we are examining metal vapor plumes produced by laser ablation of thin metal films. Such absorbing films are coated onto the back surface of transparent glass slides.

With intense, ultra-short pulses it is critical to understand the ionization process by which plasmas are abruptly formed within a single pulse. It is equally critical to understand propagation characteristics of such pulses in these plasmas (including preformed cases). We are conducting a series of experiments to examine both the intrinsic ionization products of strong OFI at collisionless gas target densities and the formation of suitably dense metal vapor plumes based on laser-induced ablation of thin metal films.

This report outlines the 'thin film' experiments in general which were conducted using both the Physics Research Laser (PRL) Facility and the Intense, Ultra-Short Pulse Laser (USP) Facility at the Lawrence Livermore National Laboratory. Following a general outline of the thin film ablation method and experimental setup is a basic description of the simultaneous, two-frequency, interferometry. Experimental results are subsequently presented from the 100 picosecond probe and the intense 125 femtosecond pulses. Rapid ionization is assumed to proceed via the strong optical field process which proceeds sequentially for the most part. It is briefly discussed prior to the summary.

3. PRODUCTION OF METAL VAPOR PLUMES BY LASER ABLATION OF THIN FILMS

The basic technique employed in this study has been described earlier by Mayer and Busch⁵ and later used in strongly coupled plasma experiments by Mostovych et al⁶. The essential geometry is outlined in Fig. 1. An adequately intense laser pulse irradiates the back surface of an absorbing metal film. This film may also be overcoated with another film depending on target requirements.

Figure 1 Target configuration for ablation studies.

After the 'formation' laser pulse reaches the absorbing film (at a time defined to be zero) the ablated material freely expands into vacuum away from the slide surface (target chamber pressure was maintained at the 10^{-5} torr level). A simple one dimensional model for this process is discussed by Mayer and Busch. For the 1 D case, the velocity of the outermost vapor edge at distance, L is the edge velocity, v_{edge} . The material density reduction with respect to initial solid density is modelled as a simple linear decomposition. The particle density, $n(t)$ temperature, $T(x, t)$ and velocity, $V(x, t)$ are written in terms of distance, x and time, t as:

$$n(t) = n_0 \left(\frac{L_0}{L} \right)$$

$$T(x, t) = T_0 \left(\frac{L_0}{L} \right)^2 \left\{ 1 - \left(\frac{x}{L} \right)^2 \right\}$$

$$V(x, t) = \left(\frac{x}{L} \right) \frac{dL}{dt} = \left(\frac{x}{L} \right) v_{\text{edge}}$$

$$v_{\text{edge}, \infty} = \sqrt{\frac{6T_0}{m_i}} \quad (2)$$

(where $0 < x < L$, $L_0 = L(0)$ and $T_0 = T(0, 0)$)

Described in this report are results from irradiation of targets comprised of a 0.5 micron layer of aluminum (167 skin depths w.r.t. intensity) overcoated with a 0.1 micron layer of carbon (1 skin depth w.r.t. intensity). The formation pulse spotsize at the aluminum layer was typically near 1 millimeter (from 0.5 to 2.0 mm) which means that approximately 3×10^{16} target particles were contained within the cylinder of diameter equal to the spotsize and length, L_0 (see fig.1) equal to 0.6 microns (0.5 + 0.1 microns). Vapor expansion according to equations (1) above suggests that when L has expanded to 0.6mm, the number density drops by three orders of magnitude below that of the initial solid layer.

The formation pulses used were of wavelength 1064nm. Although a variety of pulse widths can be used with this technique⁵ we report here on results from 8 nanosecond FWHM pulses from a commercially available Nd:YAG laser. Incident pulse energies ranged from 10's to 100's of millijoules corresponding to an on target intensity range, 10^8 to 10^{10}

W/cm^2 . It is important to remain below any threshold for glass substrate damage. The onset of strong absorption in the glass substrate has been seen to greatly diminish the amount of material ablated from the surface due to energy loss.

Figure 1 also indicates the direction of a 100 picosecond probe pulse traversing the vapor plume. The thickness of material sampled is determined by the masking used. In our case the mask consisted of a slit with width either 100 or 250 microns. The short probe pulse contained both 1064 and 532 nm laser light and could be continuously delayed relative to the arrival time of the formation pulse, typically by 10's to 100's of nanoseconds. As will be seen, the vapor plume moves about 1 micron during this 100 picosecond sampling interval. Interferograms were obtained for single laser shots using the two wavelength self-referencing setup shown in fig. 2. Infrared images were recorded using a Photometrics CCD 200 system and green images were captured from a Hitachi video camera using a framegrabber.

Fig.2 Experimental configuration for probe absorption and two-frequency interferometry of vapor plumes.

Figure 2 outlines the general experimental setup showing also the polarization splitting with the 100 psec probe. In addition to the two color interferometric feature we distinguished this larger scale P polarized interferometry from scattering of focussed S polarized 532 nm light. The position of the focussed probe could be located by superimposing part of this light onto the P polarized interferogram. This application of polarization filtering had been used earlier by Mostovych et al⁶. The ratio of sampled incident and transmitted S polarized 532 nm energy readings from Molelectron J3 sensors provided a measure of the scattering and absorption of the probe which was focussed to about 300 microns.

4. SIMULTANEOUS TWO WAVELENGTH OPTICAL INTERFEROMETRY

The sensitivity and the sign of optical phase shifting in an interferometer differs for free electrons and neutral particles for a given probe wavelength and integration length. Simultaneous utilization of two significantly different wavelengths enables distinction of electron and neutral particle densities for a single shot. The optical phase shift experienced by a probe of frequency, ω propagating through an electron density profile of physical extent, L is approximately given by⁷:

$$\Delta\phi_e \approx (L\bar{n}_e) \left(\frac{e^2}{2 m_e c \epsilon_0} \right) \left(\frac{\omega}{\omega^2 + \nu^2} \right) \approx 5.29 \times 10^{-6} \left(\frac{L\bar{n}_e}{\omega} \right) \quad (3)$$

where $\bar{n}_e$ refers to the average electron density over the profile of extent, L. Similarly the phase shift experienced by the same probe frequency propagating through a neutral density profile of equal extent is approximately:

$$\Delta\phi_n \approx L\bar{n}_n \left(\frac{\alpha}{2c\epsilon_0} \right) \omega \approx 1.9 \times 10^{-37} L\bar{n}_n \omega \quad (4)$$

where $\bar{n}_n$ refers to the average neutral density over the length, L as before. For these expressions e is the electron charge, m_e is the electron rest mass, C is the speed of light in vacuum, ϵ_0 is the vacuum permittivity, α is the neutral particle polarizability, and V represents the effective (i.e. energy distribution averaged) collision rate for momentum alteration of electrons. For a given single probe frequency the comparison of sensitivities is therefore:

$$\frac{\Delta\phi_e}{\Delta\phi_n} \approx 2.78 \times 10^{31} \frac{\bar{n}_e}{\bar{n}_n} \frac{1}{\omega^2} \quad (5)$$

By expressing phase shifts for 1064 and 532 nm wavelengths as the sum of parts induced by free electrons and neutral particles, and using in the process the frequency scaling given in equations (2) and (3), we can isolate the electronic and neutral contributions as follows:

$$\Delta\phi_e(532 \text{ nm}) = \frac{-2}{3} \Delta\phi_{1064} + \frac{1}{3} \Delta\phi_{532} \quad \text{and} \quad (6)$$

$$\Delta\phi_n(532 \text{ nm}) = \frac{-2}{3} \Delta\phi_{1064} + \frac{4}{3} \Delta\phi_{532}$$

Neutral and electron densities given below are based on the application of equation (6).

5. RESULTS USING 100 PICOSECOND PROBE PULSES

A typical shadowgraph obtained with 532 nm light is shown in fig.3. Shown in the figure is a 100 micron diameter slit placed in front of the coated surface to reduce the probed optical thickness. The leading (rightmost) edge of the shadow is seen for a probe pulse delay of 148 nanoseconds relative to the arrival time of the formation pulse at the target. According to the length scale the material the edge velocity is 1.2×10^6 cm/sec. Using equations (2) the edge velocity after sufficiently long delay times is related to the surface temperature at time zero, T_0 . This velocity suggests a surface temperature near 6 eV.

Therefore assuming the entire film thickness to be uniformly heated near time zero, 3×10^{16} irradiated particles convert approximately 30 mJ of incident laser pulse energy to directed kinetic energy. Note also that as expected there is a vapor of significantly greater optical thickness between the slit mask and the glass surface.

Fig.3. 532 nm shadowgraph with 100 micron slit.

Fig. 4. 532 nm interferogram without slit.

Fig. 5 1064 nm interferogram without slit.

Figures 4 and 5 show typical interferograms obtained with the 532 and 1064 nm wavelengths respectively for the same formation pulse. The location of the focussed S polarized probe is clear near the image center. Because the probe delay time is only 80 nanoseconds most of the focussed probe energy is transmitted through the plume. Because no masking slit was used in this case the propagation length for interferometry is of order 1 mm (i.e. a formation pulse spotsize). At early times the ablated spotsize is clearly outlined as is the limited extent of the vapor plume. For this shot the 1064 nm image (fig.5) shows clearly the stronger effect of 'free' electrons in the form of reduced fringe shifting toward the right. That is, for the displayed images free electrons induce a leftward shift while neutral particles induce a rightward shift. A spot check of both images at the referenced position, P indicates that the neutral density is of order $10^{19} / \text{cm}^3$ and the electron density is of order $10^{17} / \text{cm}^3$. Position, P is located 0.5 mm in front of the slide surface and this result is consistent with the linear decompression described in equations (2). The neutral density grows linearly as position P is moved closer to the glass slide surface.

Fig.6 Transmission for 100 picosecond 532 nm pulses through the 0.5micron Al+ 0.1micron C vapor plume.

Fig.7. Transmission of intense 125 femtosecond 800 nm pulses at 30 mJoules energy through the 0.5micron Al+ 0.1micron C vapor plume.

The S polarized probe transmission measurements were made with varying probe delay time. Results are seen in figure 6 for the probe focussed 1.2 mm in front of the slide surface and using no masking slit. The ablation pulse width is 8 nanoseconds and the target is as shown in fig.1. The transmission drops rapidly to 50% between 80 and 100 nanoseconds for a formation pulse energy of 0.4 J. This is also the case for higher pulse energies and is consistent with the 1.2×10^6 cm/sec edge velocity. In this plot there is generally higher probe transmission (> 83%) with any minima delayed later in time (> 300 nanoseconds) for much lower pulse energies. A simple explanation is that for these cases less material expands from the slide surface and at lower kinetic energies.

As indicated in fig. 1 a glass witness plate was used for some targets to collect redeposited metal. Visual observations show that most of the ablated metal is redeposited onto the second slide within a diameter of 1 cm at a distance of 14 cm from the target slide. We estimate the divergence of the emitted vapor plume to be 4 degrees.

6. RESULTS USING INTENSE, ULTRA-SHORT LASER PULSES

In a series of formation pulse shots at the 0.4 J level we replaced the 100 picosecond probe pulse with a 125 femtosecond intense pulse of central wavelength 800 nm using a configuration similar to that in figure 2. For this part of the work the experiments were conducted using the Intense, Ultra-Short Pulse (USP) Laser Facility in L-Division of this laboratory. This newer system is based on Ti:Sapphire technology. Focussing the 800 nm pulse was accomplished using a coated f/5 off-axis parabola inside a large vacuum chamber. Single pulse energy transmission was measured using Molectron J50 LP sensors to sample a portion of incident and transmitted light.

Figure 7 contains a plot of the transmitted energy fraction for ultra-short pulses of incident energy 30 mJ. Laser pulses were focussed to a spotsize of 5 microns in vacuum at a location corresponding to the vapor plume center. Plume center is well defined since a 250 micron slit mask was also placed in front of the slide target. With respect to vacuum incident peak intensities were therefore at the 10^{18} W/cm² level. An f/3 lens was used to collect transmitted light.

As seen in the figure, for intense laser pulses focussed 700 microns in front of the glass slide surface transmission is reduced to the 50% level for pulse delay times above 100 nanoseconds. This result is similar to that obtained with the longer

532 nm probe where no masking slit was used with the plume and is consistent with an edge velocity of order 10^6 cm/sec. As will be discussed later, almost all of the remaining 50% of the ultra-short laser pulse energy is scattered off-axis within the vacuum chamber. Similar to discussions presented by Monot et al⁸, we attribute the loss in transmission mostly to refractive effects associated with ionization with negligible losses due to reflection and absorption.

The intense laser pulse can induce significant ionization of the vapor plume within the 125 femtosecond interval. As will be discussed later, ionization proceeds via optical tunneling or strong optical field ionization (OFI) and is assumed to do so sequentially at this point. Interferometry was not employed for these shots, but we did observe the characteristic 'blue' spectral shift of the transmitted pulse energy. This shift is attributed to the rapid reduction of the refractive index associated with the rapid increase in the 'free' electron density due to ionization.

A simple 1 dimensional model has been presented by Penetrante et al⁹ to describe similar spectral observations with intense, 620 nm pulses of 100 femtosecond duration. For this 1 D case the spectral blue shift experienced by an ionizing pulse is written as the line integral (along the ionization length, L) of the refractive index time derivative:

$$\frac{\Delta\lambda}{\lambda_0} \approx \frac{1}{c} \int_0^L n(\dot{x}, t) dx; \quad (7)$$

$$\text{where } n \approx \sqrt{1 - \frac{\left(\frac{\omega_p}{\omega_0}\right)^2}{(1 + \kappa^2)}}$$

Here $\left(\frac{\omega_p}{\omega_0}\right)$ is the ratio of the plasma to laser (angular) frequency, c is the speed of light in the unionized medium, and κ is the average collisionality in the plasma. Equation (7) is easily rewritten in terms of the line averaged temporal derivative of the electron density, $\overline{\dot{n}_e}$ as :

$$\frac{\Delta\lambda}{\lambda_0} \approx \frac{-(e\lambda_0)^2}{8\pi^2 m_e \epsilon_0 c^3} \left(\frac{L \overline{\dot{n}_e}}{1 + \kappa} \right) \quad (8)$$

$$= -1.5 \times 10^{-24} \lambda_0^2 L \overline{\dot{n}_e}$$

where MKS units are used. We observe a maximum blue shift of about 6 nm for 30 to 50 mJ pulse energies. According to equation (8) above, $L \overline{\dot{n}_e}$ is therefore equal to $2 \times 10^{34} / \text{m}^2 \text{ sec}$ for our transmission measurements and using 250 microns for L, $\overline{\dot{n}_e}$ is of order $10^{32} / \text{cm}^3 \text{ sec}$. This estimation yields a length and time averaged $\langle \Delta n_e \rangle$ of $10^{19} / \text{cm}^3$ over the 125 femtosecond FWHM. Locally (in space and time) the increase in electron density can be much higher than this doubly averaged estimate. Precise knowledge of the electron density profile is needed to determine our proximity to the often quoted critical power, P_c for the onset of relativistic self-focusing^{3,10} :

$$P_c \approx 1.6 \times 10^{10} \left(\frac{\omega_c}{\omega_p} \right)^2 \text{ Watts} \quad (9)$$

These estimates of electron density made from spectral shifting can be compared with those made from our studies of strong optical field ionization.

7. INTERPRETATIONS FROM STRONG OPTICAL FIELD IONIZATION

Significant work is now being done to understand and distinguish the optical field ionization (OFI) mechanism from conventional multiphoton ionization (MPI)¹¹⁻¹⁹. Typically ionization can be described as a field or tunneling process when the ionization rate exceeds the laser frequency (i.e. the Keldysh parameter is well below one). OFI is clearly isolated in the long wavelength limit. For optical wavelengths ultra-short pulse durations afford improved distinction. Many laboratories now have ultra-short (of order 100 femtosecond) laser pulses capable of focussed peak intensities corresponding to many atomic units of electric field amplitude.

We had previously investigated nonresonant OFI assuming it to proceed sequentially from unperturbed neutral atom and ion ground states¹⁸. Although our more recent results from He and Ne indicate that a nonsequential fraction may occur^{12,20}

we assume here that most of the ionization is sequential in nature and that the peak laser intensity dependent ion yields follow the 'characteristic' sequential predictions. Our closed form approximations to sequential field ionization yield the following expressions for the threshold and saturation intensities, I_{th}^k and I_{sat}^k respectively, for production of a charge state, k using linearly polarized laser light :

$$I_{th}^k = 5.89 \times 10^{-4} \left(\frac{E_{k-1}^3 \epsilon_{au}^2}{\left(\ln \left(\frac{\alpha \omega_0 E_{k-1} \left(\frac{2\tau}{1.76} \right) \sqrt{\frac{\pi}{2}}}{0.01} \right) \right)^2} \right) \quad (10)$$

$$I_{sat}^k = 5.89 \times 10^{-4} \left(\frac{E_k^3 \epsilon_{au}^2}{\left(\ln \left(\frac{\alpha \omega_0 E_k \left(\frac{2\tau}{1.76} \right) \sqrt{\frac{\pi}{2}}}{4.605} \right) \right)^2} \right)$$

where α is an ionization rate constant, ω_0 is the atomic unit of frequency ($4.16 \times 10^{16} \text{ sec}^{-1}$), τ is the FWHM of the laser pulse, ϵ_{au}^2 is the square of the atomic unit of field ($2.6 \times 10^{19} \text{ V}^2/\text{cm}^2$), and E_k^3 is the cube of the ground state binding energy or ionization potential for charge state k in Hartrees. Results from these expressions are plotted in fig. 8 versus ionization potential.

Fig.8. Dependence of threshold and saturation laser intensities on ionization energy for linear polarization.

The transmission plot shown in fig. 7 suggests that at least 20% of the incident laser energy is still confined within the diffraction limited divergence. That is, a peak intensity near $3.5 \times 10^{17} \text{ W/cm}^2$ is transmitted through the diffraction limited focus. Comparison with fig. 8 shows that this is adequate to reach saturation for Al^{+5} and for C^{+3} at the focus based on sequential OFI. At a distance along the laser propagation direction equal to the confocal parameter of 142 microns the peak laser intensity drops by half to $1.8 \times 10^{17} \text{ W/cm}^2$. At this extent saturation occurs up to charge state Al^{+3} and C^{+3} . These figures indicate that along a cylinder of length equal to the 250 micron slit width the photoelectron density can range from 3 to $5 \times 10^{19} / \text{cm}^3$. This is understandably higher than the doubly averaged result of $10^{19} / \text{cm}^3$ based only on a 1D model of spectral blue shifting.

According to equation (10) the critical peak power required for the onset of relativistic self-focusing at these electron densities ranges from 5.8 to 9.8×10^{11} Watts for densities 5 to $3 \times 10^{19} / \text{cm}^3$ respectively. This exceeds our presently available peak power of 4×10^{11} Watts using 50 mJ pulse energies.

Estimates can now be made for reflected and absorbed laser pulse energy in the vapor plumes. Laser pulse energy required to induce such electron densities along a 250 micron length of vapor plume of neutral density near $10^{19} / \text{cm}^3$ is at the 10 microJoule level. Assuming an electron density of $3 \times 10^{19} / \text{cm}^3$ with an infinite gradient at the plume boundary we can calculate an energy reflectivity of order 2×10^{-5} with the refractive index given in equation (7). With reference to fig. 7 we therefore conclude that most of the energy lost in transmission is refractively scattered off-axis.

8. SUMMARY AND DISCUSSION

With 100 picosecond interferometry and absorption we have demonstrated that laser ablation of thin metal films is a viable technique for production of dense vapor plume targets to be used in a variety of laser plasma interaction experiments. Based on both S polarized absorption measurements and shadowgraphy the metal vapors are emitted with speeds of order 10^6 cm/sec (or 1 micron/ 100 picoseconds), are relatively well directed, and can be masked to produce desired density profiles and spatial extents. For a specific slide target configuration the principal controls on vapor plume characteristics are formation pulse energy and delay time relative to the arrival time of the formation pulse at the thin film. For the thin film coating discussed in this report vapor densities greater than $10^{19} / \text{cm}^3$ occur closer to the target slide surface (i.e. closer than 0.5 mm). Furthermore, thicker films can be deposited onto glass slides to ablate more material. Tight focusing requirements imposed by highest achievable peak intensities for laser pulses can also place restrictions on glass slide width.

Transmitted energy fractions and spectra were recorded with intense, ultra-short pulse irradiation of these vapor plumes. Temporally and spatially averaged estimates of induced electron density in the plume were made by applying a simple 1D model for spectral blue shifting. In addition energy transmission observations were combined with sequential OFI theory to infer electron densities expectedly higher than those inferred from transmitted spectra. Most of the energy loss deduced from limited transmission is attributed to off-axis scattering associated with ionization induced refraction.

We plan to apply the thin film ablation technique as well as gas nozzle technology to laser plasma interaction experiments for intense, ultra-short pulses. Electron densities of order $10^{20} / \text{cm}^3$ are required for studies of OFI driven recombination x-ray/xuv lasers and relativistic propagation effects. Interferometry must be included to verify electron densities. Both gas nozzle and thin film target sources can be used in experiments with repetition rates of a few pulses per second. Finally, it is clear that the fundamental examination of the OFI process must be extended to highest achievable laser intensities. Interesting observations for intensities up to the 10^{16} W/cm^2 level have been reported in which a fraction of nonsequential OFI or double ionization in He is shown to predict ion yields^{12,20}. Studies of the effects of laser pulse wavelength and polarization on intensity dependent ion yields for high charge states are underway for intensities up to 10^{18} W/cm^2 .

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glass
substrate

100 micron
slit mask

0.5Al + 0.1C (microns)
148 ns delay
532 nm shadowgraph

(0.5 mm length)

plume edge
at $x=L=1.8$ mm

glass
substrate

$0.5\text{Al}+0.1\text{C}$ (microns)
80 ns delay
1064 nm

—▶
P

S Polarized 532 nm
Probe at 1.2 mm

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