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Imaging femtosecond laser induced electronic excitation in glass

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ABSTRACT

While substantial progress has been achieved in understanding laser ablation on the nanosecond and picosecond time scales, it remains a considerable challenge to elucidate the underlying mechanisms during femtosecond laser material interactions. We present experimental observations of electronic excitation inside wide band-gap silica glass during single femtosecond laser pulse (100 fs, 800 nm) irradiation. Using a femtosecond time-resolved imaging technique, we measured the evolution of a laser-induced electronic plasma inside the silica glass and calculated the electron number density to be on the order of 10^{19} cm^{-3}.

Pulsed laser ablation has been demonstrated to be a viable technology for an increasing number of applications. These include pulsed laser deposition of high critical-temperature superconductor films[1,2], femtosecond laser micro-machining and structural modification of dielectrics[3,4], ultrafast laser-assisted material and chemical analysis[5,6], and ultrashort X-ray pulse generation[7,8]. However, high power, short pulse laser ablation is still largely unexplored at the fundamental level.

Mechanisms underlying laser ablation processes are quite complex, most previous investigations have focused on laser ablation at relatively long (e.g., nanosecond and picosecond) time scales[9,10]. Since the femtosecond time scale is much shorter than the electron-lattice energy relaxation time, thermal equilibrium cannot be established during the time the laser interacts with a solid material. The absence of a quantitative model at least in part contributes to the lack of understanding of femtosecond laser ablation. Systematic time-resolved measurements can help establish a predictive theory for laser ablation on the femtosecond time scale. In this study, we aim to elucidate some of the characteristics resulting from femtosecond laser interactions with a transparent solid. A detailed examination is presented of the development of laser-induced electronic exciation inside silica glass on the femtosecond time scale. An electronic plasma generated by a 100 fs, 800 nm laser incident in the silica glass was measured.

Laser ablation experiments were performed using a femtosecond time-resolved pump-probe setup to image the plasma. Details of the experimental setup will be published elsewhere. Briefly, a Spectra-Physics TSA high power femtosecond laser at its

fundamental wavelength (800 nm) was used as the pump beam, which has a pulse duration of approximately 100 femtosecond (FWHM) and energy output up to 25 mJ per pulse. The 800 nm laser beam was focused to a spot size of 50 μm in diameter onto a silica glass sample using an f = 15 cm focal-length lens. After a beam splitter, one arm of the 800 nm output passes an optical delay stage and a KDP crystal, forming a probe beam at 400 nm that is perpendicular to the ablation laser beam. By moving the delay stage on an optical rail, the optical path of the probe beam can be varied, so the time difference between the ablation beam and the probe beam is changed. In all of the experiments, time zero was set when the peaks of the ablation laser beam and the probe beam overlapped in time at the sample surface. Shadowgraph images inside the silica were obtained by measuring spatial transmittance during laser irradiation of the sample and correcting for background intensity measured without laser ablation. Shadowgraphs were recorded with a CCD camera (Photometrics AT200). Electron number density of the laser-induced plasma inside the silica sample was estimated from the transmittance images at various delay times.

Laser induced filamentation, as well as the related nonlinear self-focusing processes[11], have been investigated for decades (e.g., femtosecond laser filamentation in air[12] and in dielectrics[13]). However, there are few femtosecond time-resolved studies of laser self-focusing and filamentation inside solid media. Figure 1 shows a series of shadowgraph images taken at the same delay time (2000 fs) but at different laser irradiance I. The silica glass-sample surface can be seen clearly in this figure, which separates the silica glass on the left-hand side and the air to the right. Weak fringes close to the sample surface are due to edge diffraction of the probe laser beam. At $I = 5 \times 10^{12}$

W/cm^2, there is only one filament, a thin, dark stripe which results from the absorption of the probe beam by laser-excited electrons inside the silica glass. The strongest absorption at this laser irradiance appears to locate a few hundred μm away from (inside) the glass surface. This location is a direct result of the propagation of the 100 fs femtosecond ablation laser pulse (30 μm FWHM spatial extent), whose peak is a few hundred μm inside the glass after the experimental delay time. The absence of a dark filament close to the glass surface at $I = 5 \times 10^{12}$ W/cm^2 is due to the relaxation of laser-excited electrons; the resulting electron number density is not high enough to cause significant absorption of the probe beam. With increased laser irradiance ($I = 1.3 \times 10^{13}$ W/cm^2), more electrons are excited and therefore, the filaments appear longer than those at low irradiances.

At high laser irradiance, the filament splits. At $I = 2.5 \times 10^{13}$ W/cm^2 (Fig. 1), the primary filament splits into two at a location about 200 μm inside the silica glass. At even higher irradiance (e.g., 10^{14} W/cm^2), splitting starts right after the femtosecond laser pulse enters the glass sample. Other than filament splitting, figures 1 shows the evidence of strong self-focusing of a femtosecond laser pulse inside silica glasses. The width of the filaments appears to decrease as the laser pulse propagates into the sample, especially at high laser irradiances.

Experiments were performed to measure the electron density inside the femtosecond laser-excited silica glass. Figure 2 shows a series of time-resolved images of the plasma filaments at the same laser irradiation $I = 1.3 \times 10^{13}$ W/cm^2 (one filament). At $t = 0$, only a small dark area appears close to the glass surface that results from

electron excitation by the rising edge of the femtosecond laser pulse. At longer delay times, plasma filaments grow longer, with the darkest section (strongest absorption) moving away from the glass surface, reflecting femtosecond pulse propagation. From the transmittance data of these time-resolved images, we can estimate the femtosecond laser-excited plasma electron number density inside the silica glass at different delay times.

Let I_p represent the probe (400 nm) laser pulse intensity, α the absorption coefficient of the plasma inside the glass, and z the spatial dimension with $z = 0$ set at the glass surface. According to the differential expression (one-dimensional), $dI_p/dz = -\alpha I$, we can determine the absorption coefficient as, $\alpha = 1/d \ln(I_{p0}/I_{pd})$, where d is the mean diameter of the filament, and I_{p0} and I_{pd} are probe laser intensities before and after passing through the laser-irradiated glass sample. The absorption coefficient is directly related to the plasma frequency ω_p,

$$\alpha = \frac{\tau}{nc} \frac{\omega_p^2}{1 + \omega^2 \tau^2} ,$$

where n is the refractive index of the silica glass, c the light speed, ω the probe beam light frequency, and τ the scattering time chosen as 0.2 fs. The plasma frequency ω_p is a function of electron number density n_e, as well as the electrical permissivity, and the mass and charge of a single electron. From the above analysis, electron number density n_e may be determined by measuring probe beam intensity transmittance I_{p0}/I_{pd} from the time-resolved plasma images. The above analysis provides an order-of-magnitude estimate of n_e; the three-dimensional feature of the plasma and the dependence of τ on electron number density were ignored.

The electron number density of the laser-induced plasma inside the silica glass, as shown in figure 2, was determined using the above analysis; the results are plotted in figure 3. At t = 333 fs, there is a electron number density maximum (n_e = 2 x 10^{19} cm^{-3}) at z = 80 μm; this peak moves into the glass at later times. While the peak value of the electron number density increases with time, it reaches a maximum of 5 x 10^{19} cm^{-3} at t = 1333 fs. This observation is consistent with previous reports that a femtosecond laser pulse could experience initial self-focusing inside a solid medium, followed by defocusing when the laser-induced electron excitation is strong enough to compensate the laser-induced refractive index change[11]. For comparison, an electron number density of 5 x 10^{19} cm^{-3} is about twice the standard air density.

Propagation of the femtosecond laser-induced plasma front inside the silica glass is plotted in figure 4. The data represent experimental measurements using five different excitation laser energies. The straight line in the figure is a linear fit of the experimental data. We find that the plasma propagates at a speed of approximately 1.8 x 10^8 m/s. From this measurement, we estimate that the refractive index of the silica glass is about 1.65, in good agreement with its true value (1.60). Additionally, this linear relation provides evidence that spatial evolution of the plasma electron density is a result of femtosecond laser pulse propagation inside the glass medium.

Understanding the fundamental physical mechanisms of femtosecond laser induced excitation in transparent solids could benefit the rapid development of photonic

crystal and optical storage technologies[4,14]. We investigated femtosecond laser-induced electronic excitation and subsequent plasma evolution inside a transparent silica glass. High-density electron excitation and related filamentation phenomena were observed inside the femtosecond laser-irradiated glass sample. Spatial electron density profiles at various times were measured and showed a moving peak structure. The peak electron number density increased initially, but stopped increasing after the femtosecond laser pulse propagated about 200 μm into the glass. These data suggest a defocusing process after an initial self-focusing caused by the excitation of a high-density electron plasma.

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Figure Captions:

Figure 1. Irradiance dependence of femtosecond laser plasma inside silica glass.

Figure 2. Time-resolved images of femtosecond laser-induced plasma inside silica glass.

Figure 3. Evolution of electron number density profile inside a femtosecond laser-irradiated silica glass.

Figure 4. Propagation of the femtosecond laser-induced plasma front inside a silica glass.

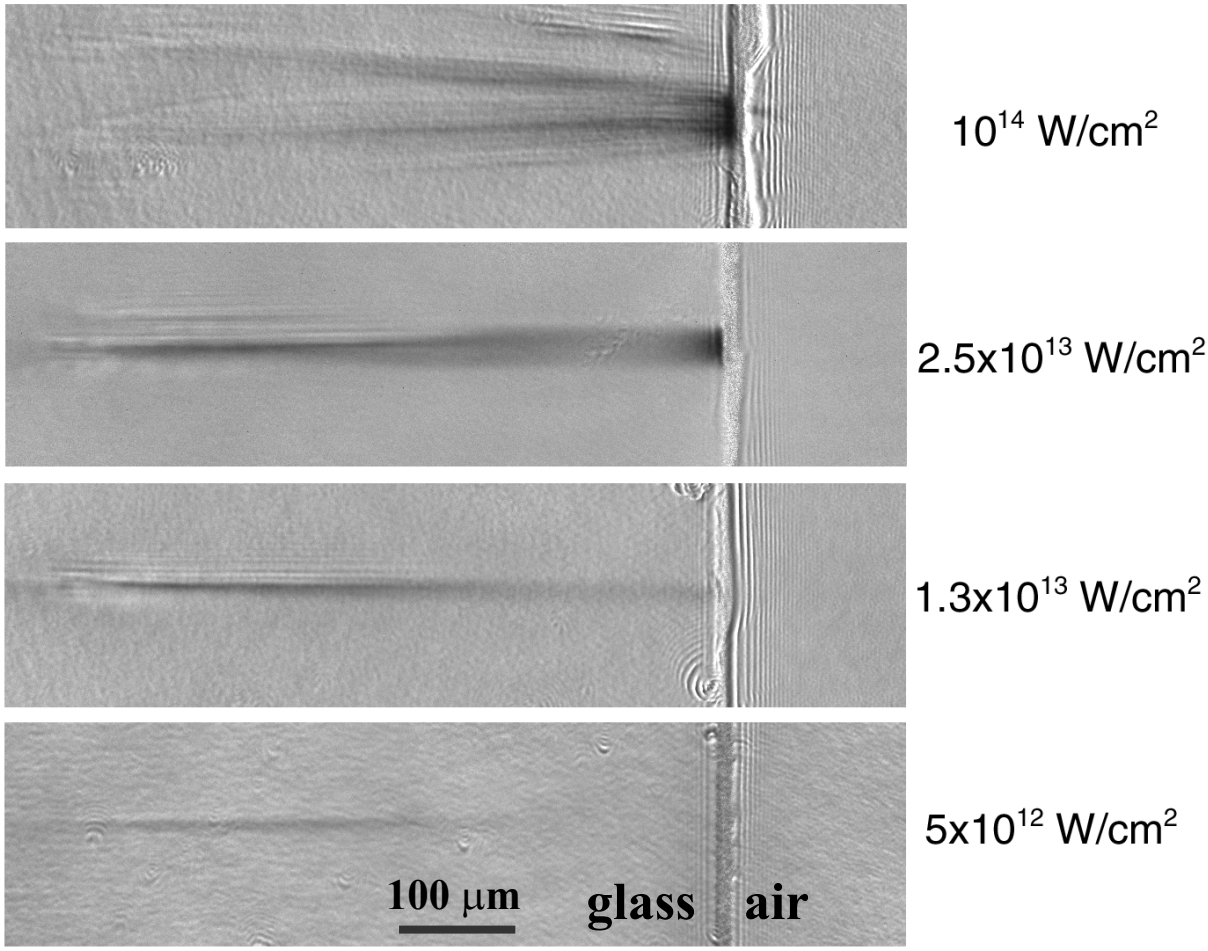
10^{14} W/cm^2
2.5x10^{13} W/cm^2
1.3x10^{13} W/cm^2
5x10^{12} W/cm^2
100 μm
glass
air

Figure 1

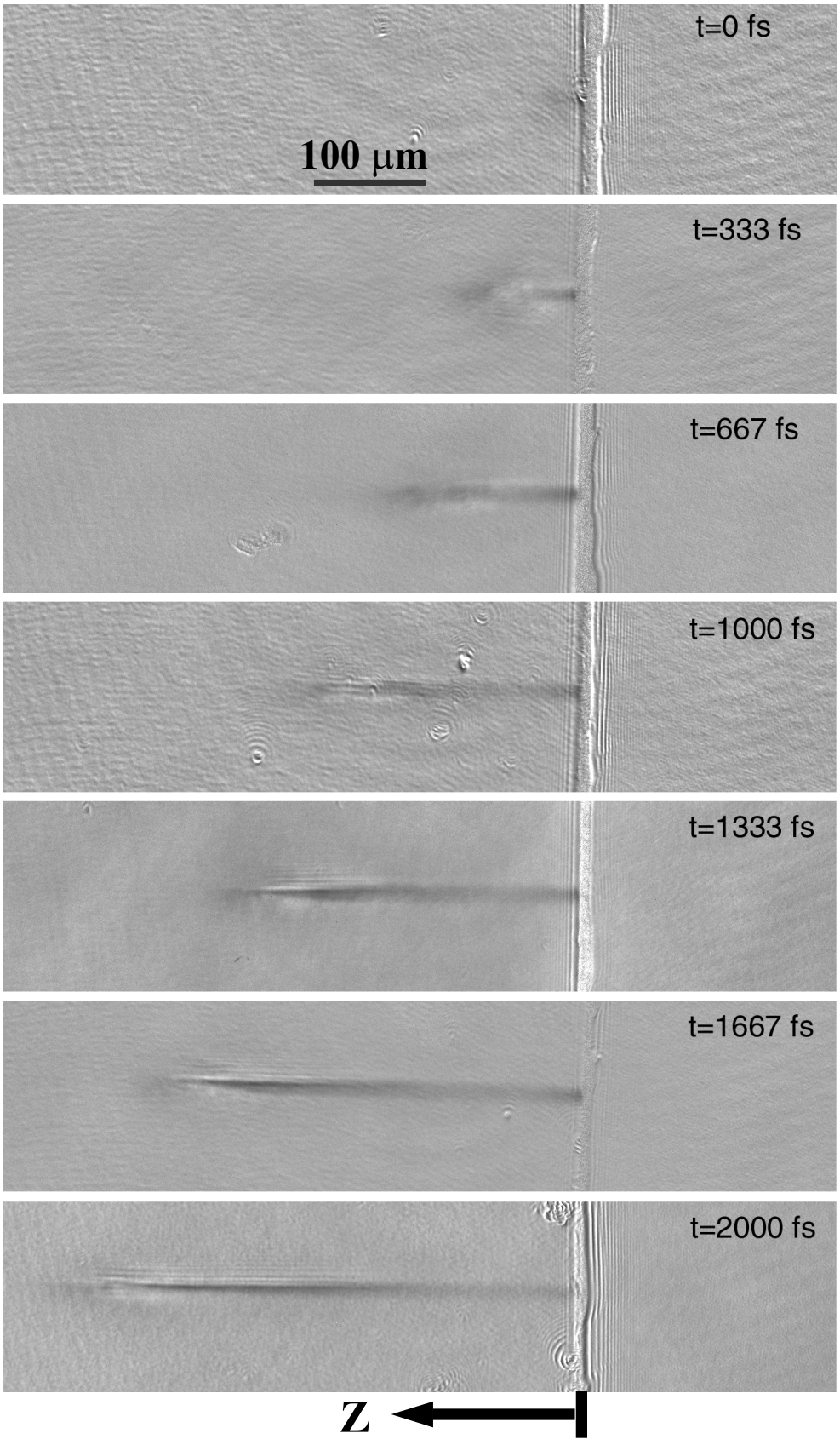
t=0 fs
100 μm
t=333 fs
t=667 fs
t=1000 fs
t=1333 fs
t=1667 fs
t=2000 fs
Z

Figure 2

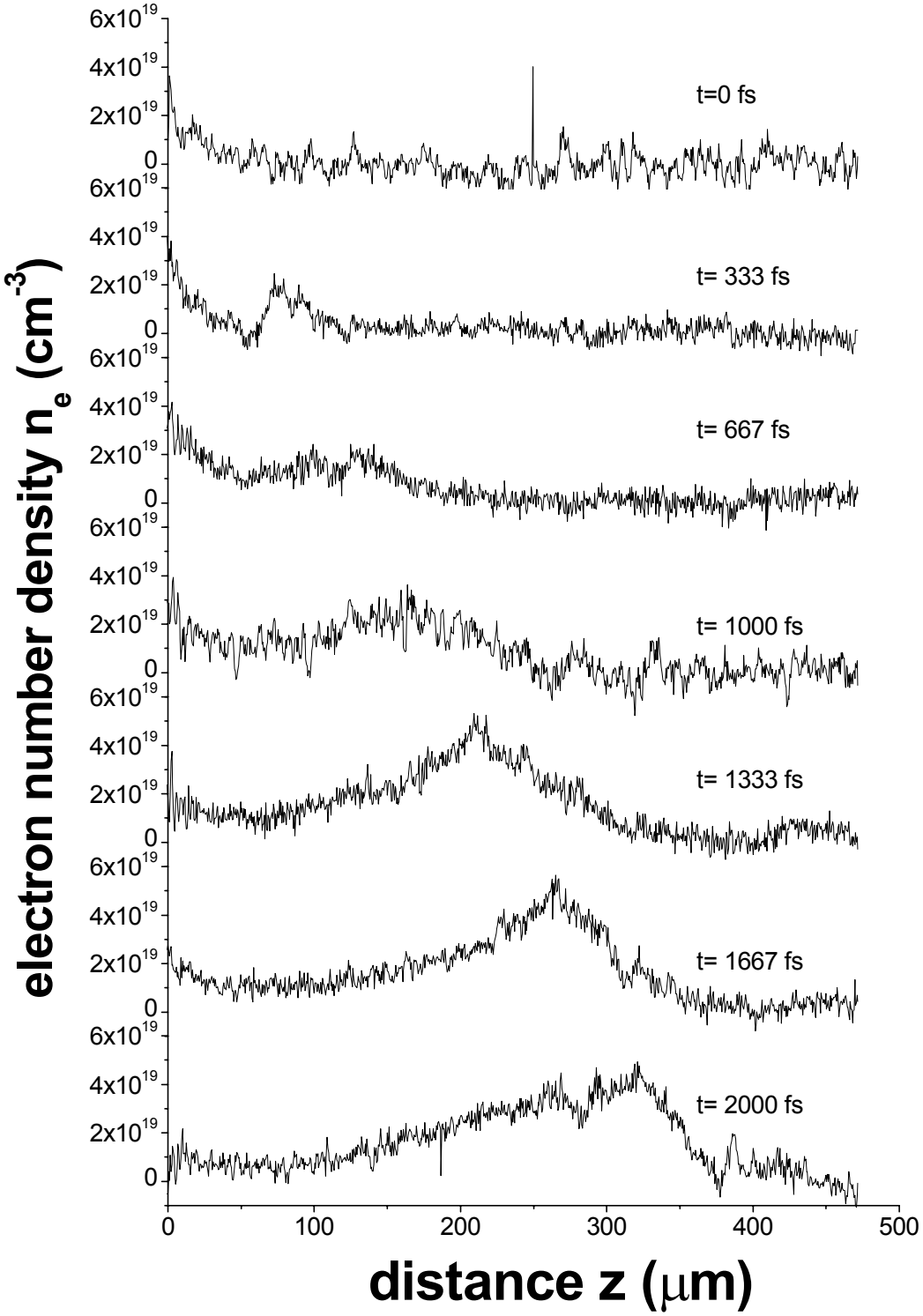
electron number density n_e (cm^{-3})
6x10^19
4x10^19
2x10^19
0
t=0 fs
t= 333 fs
t= 667 fs
t= 1000 fs
t= 1333 fs
t= 1667 fs
t= 2000 fs
0
100
200
300
400
500
distance z (μm)

Figure 3

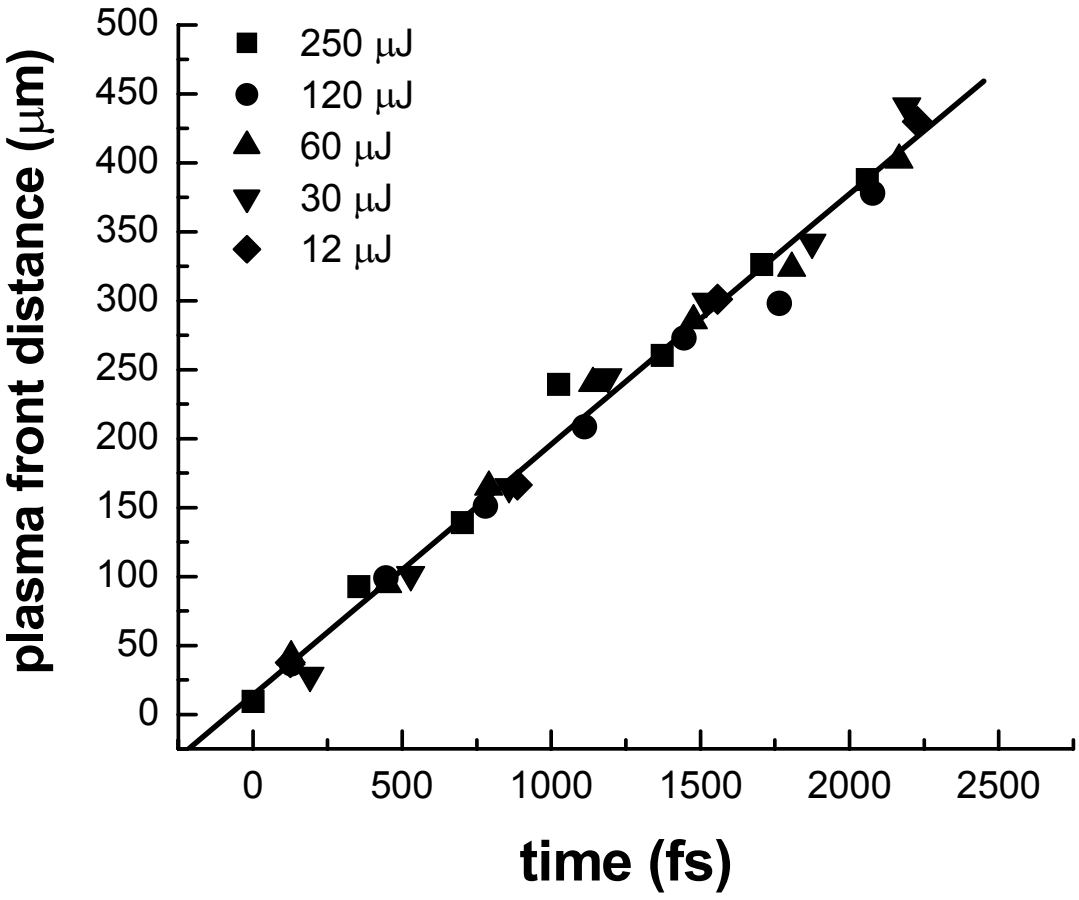
plasma front distance (μm)
time (fs)
250 μJ
120 μJ
60 μJ
30 μJ
12 μJ
0
50
100
150
200
250
300
350
400
450
500
0
500
1000
1500
2000
2500

Figure 4