Ultrafast non-linear optical method for generation of flat-top shocks

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ABSTRACT

Flat top shocks generated reproducibly by short pulse lasers are useful in studies of shock compression phenomena and may have applications in materials science, biology, and medicine. We have found the fluence profiles of Gaussian spatial mode 120-400 fs duration incident laser pulses are reproducibly flattened via surface optical breakdown in dielectric substrates at fluences just above the breakdown threshold. These flat top laser profiles have been used to produce shocks flat to 0.7 nm RMS over a 75 – 100 μm diameter.

Keywords: Ultrafast lasers, non-linear optics, self-focusing, self-phase modulation, laser-driven shocks

1. INTRODUCTION

To ensure one-dimensional stress in model shock wave systems, extreme tolerances on projectile flatness and tilt, as well as target flatness, are necessary. In the case of laser-driven shock waves, this need translates into a requirement of a uniform laser fluence distribution over the spatial region of interest. Here we demonstrate a simple method for producing shocks with very high spatial uniformity by utilizing ultrafast laser ablation as a pulse shaping mechanism.

Optical breakdown in dielectric materials using ultrashort pulses is an active area of research, especially because of its importance in ablation and micro-structuring .[1-5] The recent availability of sub-picosecond laser systems has enabled micro-structuring with sub-micrometer resolution because such short laser pulses avoid the pitfalls of strong thermal effects and plasma etching that occur with nanosecond laser sources. Studies of the laser/material interaction mechanism in the sub-picosecond time regime have resulted in several findings of significance. Ablation from ultrashort pulses was found to be deterministic instead of statistical.[4] The damage threshold fluence was found to be higher than predicted from the $t^{1/2}$ scaling law for pulses < 10 ps, in spite of the expectation that multi-photon ionization or other nonlinear effects would reduce the damage threshold for shorter and shorter pulses. These recent studies[1-5] show that the damage can be associated with rapid buildup of conduction electrons to a critical density necessary for further absorption of laser energy. With shorter and shorter pulses, electrons generated by multi-photon ionization or tunnel ionization become more important than background carriers for development of the avalanche, but the avalanche mechanism predominates even for sub-100 fs pulses.[5]

One consequence of this optical breakdown is a very strongly non-linear absorption of the laser fluence transmitted through the dielectric material, which leads to flattening of the central peak of an incident Gaussian fluence profile. We have characterized the transmitted pulse fluence profile and have found it to be flat over a significant portion of the parameter space just above the threshold fluence (for 120-400 fs pulses). In addition, we have used the transmitted flat profile to drive flat shocks into thin film metal layers vapor deposited onto the dielectric materials and have characterized them using spatially resolved frequency domain interferometry.

2. EXPERIMENTAL

The apparatus used for generation of flat top shocks is fully described in references 6 and 7. It utilized 120-400 fs laser pulses at 800 nm from a seeded, chirped pulse amplified (CPA) Ti:sapphire laser. The pulses were focused to a nominal spot diameter of 75 μm at the sample. Samples for the shock wave studies were thin metal films of several thicknesses (from 0.05 to 2 μm) vapor plated onto dielectric substrates, usually borosilicate glass microscope slide cover slips. Samples for the transmittance and beam intensity profile studies were the same substrates with no coating. The samples were mounted on a

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computer-controlled x-y translation stage and were rastered about 4 laser focus diameters between experiments so that each laser pulse would propagate into undisturbed material.

The shock waves were characterized at the metal/air surface using the technique of reflection frequency domain interferometry[8, 9] to simultaneously measure the dynamic phase shift and reflectance between a pair of ultrafast probe pulses. Details of the experimental approach can be found in Ref. 6. A small portion of the main CPA laser pulse (~ 0.04 mJ) reflected from a beam splitter was passed through an unbalanced Michelson interferometer to produce a pair of probe pulses separated in time by 4-12 ps. These pulses were loosely focused on the backside of the target at an angle of $\theta = 32.6\,°$ to a spot size of ~ 200 μm to circumscribe the region of shock break out. In all cases, the probe intensity was less than ~ 5×10^{11} W/cm^2. The reflected probe pulses were imaged at x 16 magnification onto the entrance slit of a high resolution imaging spectrograph (Acton model 300i) with CCD detector (Photometrics model SenSys 1600). The details of the data analysis used to extract the relative phase shift between the probe pulses caused by motion of the surface or transient changes in the optical properties of the surface during shock break out are given in References 6 and 7.

Analysis of the laser pulse intensity profile was done using a beam-profiling CCD camera and analysis software (Spiricon). The back surface of the sample was imaged onto the CCD using an f/3 lens and x 7 magnification. An abraded portion of the sample was used to determine the correct focal plane. The camera and software were also used in conjunction with calibrated neutral density filters to determine the transmittance through the samples.

3. RESULTS AND DISCUSSION

Figure 1 shows the shock profile at the time of peak surface velocity (see reference 6 for details) for several incident laser fluences. The development of a flat shock profile is evident at fluences of 2.2 J/cm^2 (energies of 100 μJ) and above. The scales are deceptive in Fig. 1. The FWHM width of the 50 μJ trace is approximately 75 μm, but the vertical scale is phase shift $\Delta\phi$ (in radians), which can be translated to surface motion Δx using $\Delta x = \frac{\Delta\phi\lambda_0}{4\pi\cos\theta}$. For these $\lambda_0 = 800$ nm probe pulses and the 32.6 ° angle of incidence, a phase shift of 0.1 rad translates to a surface motion of only 7.5 nm or 10^{-4} of the beam diameter. The shock was flat to 0.01 rad RMS phase shift, or 0.7 nm RMS over a 75 μm diameter region.

Under the same conditions as for the 170 μJ trace of Figure 1, the intensity profile of the transmitted laser beam after passing through an uncoated cover slip is given in Figure 2. The same essential flat top features can be seen in both the intensity profile and the shock profile.

The fraction of the pulse energy transmitted by the cover slip samples as a function of incident energy and laser pulse length is given in Figure 3. To corroborate whether the flat top profile is due to optical breakdown, a simple model that assumes clipping of the intensity from the large effective absorption coefficient above breakdown was used and compared to the experimental data. The effective absorption coefficient was obtained from measurements of ablation etch rate versus incident fluence.[10] Etch rate measurements were made by exposing locations on a cover slip sample to variable numbers of pulses at a variety of pulse fluences. Etch depths were measured using profilometer and AFM techniques. Figure 4 gives the resultant etch rates d versus the natural logarithm of fluence. The line in Fig. 4 is a linear fit to the data assuming $d = 1/\alpha^* \ln(F/F_{th})$, where α^* is the effective absorption coefficient.[10] The ordinate intercept gives $F_{th} = 2.07$ J/cm^2, in good agreement with the borosilicate result in Ref. 3, and the slope gives $\alpha^* = 3.36\ \mu m^{-1}$. The transmitted energy fraction can then be written:

$$f_{tr} = \frac{2}{\sqrt{\pi}}\left\{\frac{F}{F_{th}}\sqrt{\ln(F/F_{th})} + \frac{\sqrt{\pi}}{2}\operatorname{erfc}\left[\sqrt{\ln(F/F_{th})}\right]\right\} \qquad (1)$$

The variation of F_{th} with pulse length given in Ref. 3 was used with the above equation to generate the curves given by the lines in Figure 3. This simple model accounts for the essential features of the data, although there is some discrepancy noted at large incident energies (where there may be contributions from other non-linear optical effects). Additional corroboration of this model was obtained by noting that measurements of the flat top width versus incident energy compared reasonably well to the widths of a Gaussian truncated at F/F_{th}.

To examine the relative roles of the surface ablation process and the bulk non-linear effects on the transmitted beam spatial profile, a 1 mm thick borosilicate glass sample was studied. In this thicker sample, bulk self-phase modulation and self focusing were evident at incident fluences considerably below the ablation threshold. At or above F_{th}, the transmitted profile exhibited features suggesting a combination of surface and bulk effects. At no incident fluence were we able to reproduce the quality of flatness observed in the thin samples. It is likely that the propagation distance in the thin samples is insufficient for bulk non-linear effects to play a strong role in beam shaping at the fluence levels investigated.

We also examined 1 mm thick sapphire and LiF substrates. The 1 mm LiF sample produced results similar to the 1 mm thick borosilicate glass. some flattening was evident at low fluences, but was accompanied by a ring-like intensity distribution, probably due to self-phase modulation. In the 1 mm thick sapphire sample, non-linear effects at low fluences led to excellent flattening of the transmitted beam (see Figure 5). However, the transmitted intensity was low so that it was not useful for shock generation. Higher incident fluences led to severe self-focusing and other bulk non-linear optical effects as evidenced by severe pulse shape distortion. Longer pulse lengths extended the range for flattening to higher fluences. However, pulse lengths longer than 500 fs did not achieve suitably flat profiles, possibly due to a complicated pulse shape or competition between non-linear effects.

Other substrate materials and thicknesses might be used to optimize the fluence transmitted (and therefore the shock pressure achievable). Laser pulse lengths and shapes could also be varied to optimize the flatness, reproducibility, and transmitted fluence.

4. CONCLUSION

We have demonstrated that non-linear optical phenomena in transparent dielectric materials can be utilized to produce flat top shock profiles in thin metal samples. We found the intensity profiles of incident Gaussian spatial mode 110, 210 and 400 fs duration laser pulses were reproducibly flattened via optical breakdown in thin dielectric substrates at fluences above the breakdown threshold. The flat top profiles were used to produce shocks flat to 0.7 nm RMS over diameters of 50-75 µm. Non-linear absorption induced clipping of the incident Gaussian intensity profile via a simple electron avalanche model provides a reasonable fit to transmitted versus incident pulse energy data in the borosilicate glass cover slips. Measurements of the flat top width versus incident energy and experiments in thicker substrates (BK-7, LiF, and sapphire) further support this model.

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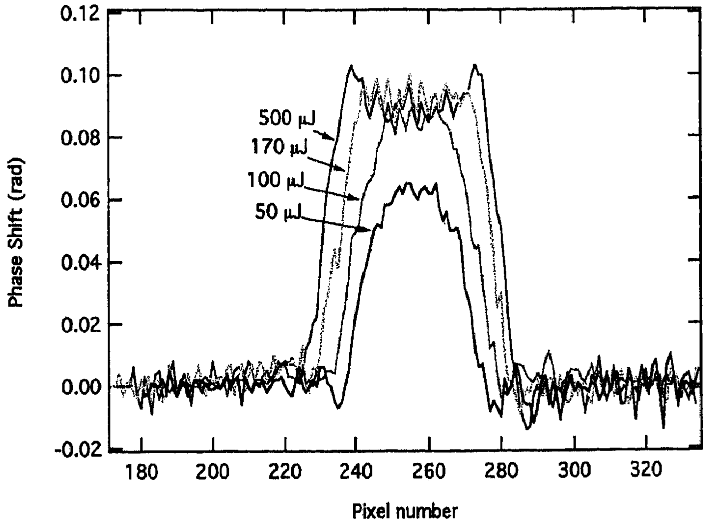

Figure 1: Shock profile at several incident laser fluences, 150 µm cover slip, 1 µm Al

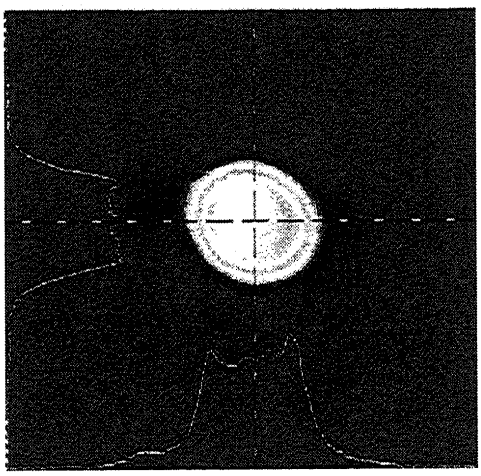

Figure 2: Image of the transmitted laser beam, 184 µJ incident energy, 150 µm thick cover slip

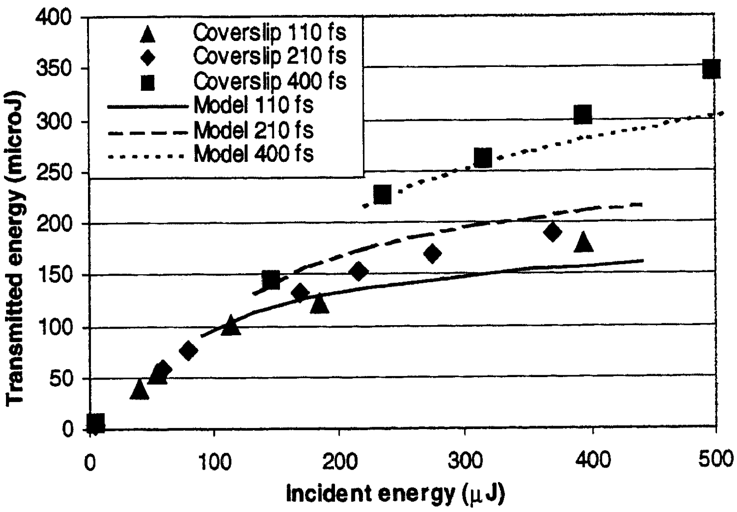

Figure 3: Transmitted versus incident pulse energy experimental data (symbols) and calculation (curves; see text).

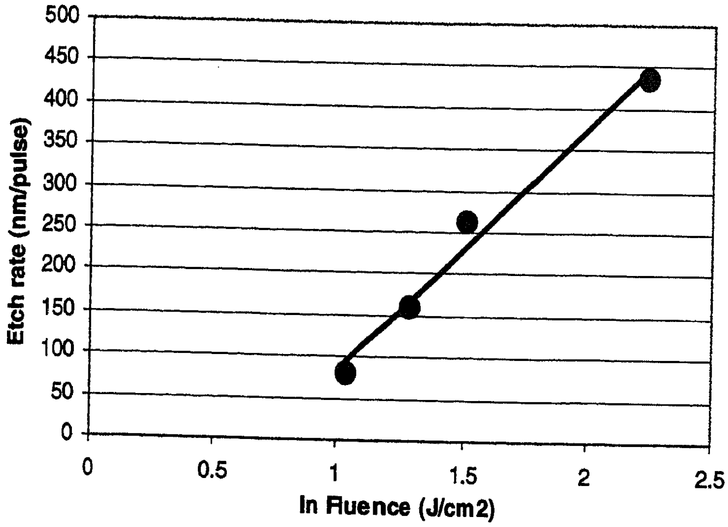

Figure 4: Etch rate versus ln incident fluence, cover slip sample, 110 fs pulse length

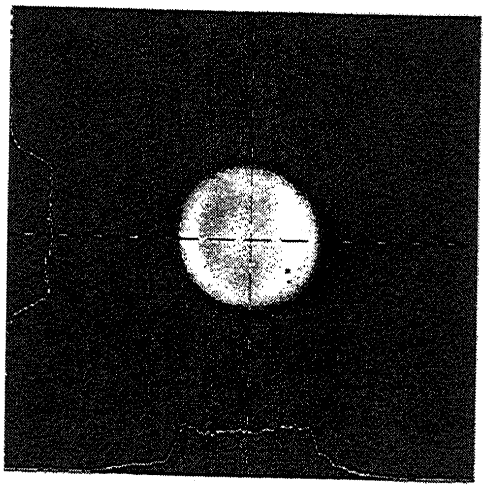

Figure 5: Image of the transmitted laser beam, 19 μJ incident energy, 1 mm thick sapphire