Ultrafast nonlinear optical method for generation of planar shocks

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Planar shocks generated 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 120–400 fs duration Gaussian spatial mode incident laser pulses are reproducibly flattened via surface optical breakdown and Kerr focusing in thin dielectric substrates at fluences just above the ablation threshold. These flat laser profiles have been used to produce planar shocks that are flat to 0.7 nm root-mean-square over a 75–100 μm diameter. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1337629]

To ensure one-dimensional strain 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 uniform laser fluence distribution over the spatial region of interest. Previous attempts to produce planar laser driven shocks have utilized optical fiber coupling.[1,2] Intra- and extra-cavity laser beam shaping methods have been used to produce flat laser fluence profiles, which could be used to drive shocks.[3,4] Here, we demonstrate a simple method for producing shocks with very high spatial uniformity by utilizing a combination of ultrafast laser ablation[5–8] and Kerr focusing[9,10] as a pulse shaping mechanism. To understand the process, we have characterized the transmitted pulse fluence profile in thin borosilicate glass samples and have found it to be flat [i.e., <5% root-mean-square (rms) intensity variation over >75% of the focal diameter] for a significant portion of the parameter space just above the ablation threshold fluence (for 120–400 fs pulses). Slightly different behavior was observed for thin sapphire and LiF samples, which can be related to differences in their ablation threshold fluences and Kerr coefficients. In addition, we have employed this effect to drive planar shocks into thin metal layers vapor deposited onto the dielectric materials and have characterized them using spatially resolved frequency domain interferometry. The short duration shocks thus produced are important for the elucidation of shock induced processes at the molecular scale.

The apparatus used for the generation of planar shocks is fully described in Ref. 11. It utilized 120–400 fs laser pulses at $\lambda_0 = 800$ nm from a seeded, chirped pulse amplified (CPA) Ti:Sapphire laser (Spectra Physics). 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 (0.05 to 2 μm thick) vapor plated onto dielectric substrates, usually borosilicate glass microscope slide cover slips (Fisher Scientific). Samples for the transmittance and beam intensity profile studies were the same substrates with no metal coating. The samples were mounted on a computer controlled *x-y* translation stage and were rastered about 4 laser focus diameters between shots 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,[12,13] using a small portion of the main CPA laser pulse reflected from a beam splitter (~0.04 mJ). Analysis of the transmitted laser pulse fluence profile was done using a beam profiling charge coupled device (CCD) camera and analysis software (both Spiricon). The back surface of the sample was imaged onto the CCD using an $f/3$ lens and ×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.

Figure 1 shows the shock profile at the delay time of peak phase shift[11] for a 1 μm thick Al film on a borosilicate cover glass (~150 μm thick) at several incident laser fluences. The development of a planar shock profile is evident at fluences of 2.2 J/cm^2 (energies of 100 μJ) and above. The phase shift $\Delta\phi$ (in radians) can be translated to surface motion Δx using $\Delta x = \Delta\phi\lambda_0/4\pi\cos\theta$. Figure 1 shows the shock is flat to 0.01 rad rms phase shift, or 0.7 nm rms, over

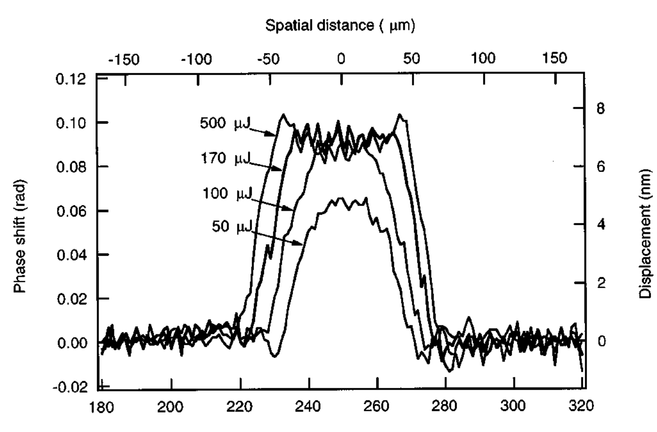

FIG. 1. Shock profile at several incident laser energies for a 1 μm Al film deposited on a 150 μm thick borosilicate glass microscope cover slip.

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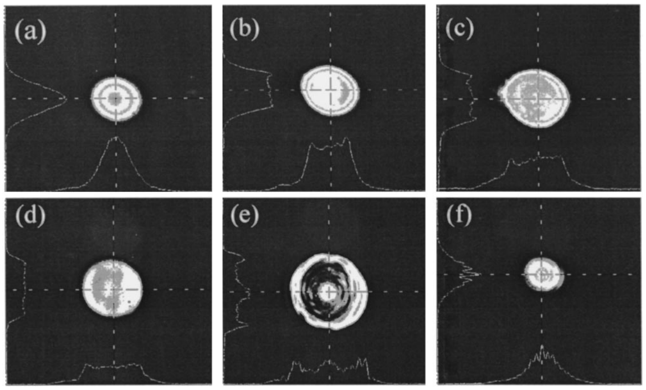

FIG. 2. Images of the transmitted laser beam: (a)–(c) 150 μm thick BK-7 cover slip: (a) 40 μJ, (b) 184 μJ, (c) 395 μJ incident pulse energy; (d)–(e) 1 mm sapphire: (d) 19 μJ, (e) 40 μJ incident pulse energy; (f) 1 mm BK-7: 7.3 μJ incident pulse energy. These are gray-scale representations of false color images.

the central 75 μm diameter region for fluences from approximately 150 to 500 μJ with pulses of 110 fs length.

Under very similar conditions as for the 170 μJ trace of Fig. 1, the intensity profile of the transmitted laser beam after passing through an uncoated cover slip is given in Fig. 2(b). The same essential flat features can be seen in both the intensity and shock profiles. At approximately 50 μJ, the shock is essentially Gaussian in shape (Fig. 1), and the transmitted beam remains Gaussian [Fig. 2(a)].

Because the observed flattening of the transmitted beam profile began at incident energies close to the ablation threshold, we considered ablation to be a possible cause of the flattening. A simple model was used to establish whether the flat transmitted beam profile could be due solely to optical breakdown. The model assumes that the large effective absorption coefficient above breakdown truncates the Gaussian incident profile. The effective absorption coefficient was obtained from measurements of ablation etch rate versus incident fluence.[14] Etch rate measurements (profilometer and atomic force microscopy) were made by exposing locations on a cover slip sample to variable numbers of pulses at a variety of pulse fluences. A linear fit of the etch rate d versus the log of the fluence F, assuming $d=1/\alpha^* \ln(F/F_{th})$ where α^* is the effective absorption coefficient and F_{th} is the fluence at the ablation threshold,[14] gave $F_{th}=2.07\,\mathrm{J/cm^2}$, in good agreement with the borosilicate results in Ref. 7, and $\alpha^*=3.36\,\mu\mathrm{m}^{-1}$.

The results of this model were compared to the transmitted versus incident energy measurements for pulse lengths of 110, 210, and 400 fs. The variation of F_{th} with pulse length was taken from Ref. 7. This simple model accounted for the essential features of the data, although it overestimated the loss at large incident energies (where contributions from Kerr focusing are significant—as will be discussed). In addition, the single pulse ablation crater diameters were only ~70% of the flat region diameter at fluences of 200 μJ and above, and were not observable at 100–120 μJ (by scanning electron microscopy),[15] where flattening in both the transmitted laser profile and the shock wave was observed in the thin borosilicate coverslip. This latter observation and the apparent onset of a ring-like structure at higher fluences [Fig. 2(c)] suggested that bulk nonlinear optical effects may play a strong role in the observed flattening process.

The role of bulk nonlinear optical effects was examined using transmitted beam profiles from 1 mm thick borosilicate glass, sapphire, and LiF samples, as well as a 100 μm thick sapphire sample. Some of these images are presented in Fig. 2. In the 1 mm samples, bulk self-phase modulation and self-focusing were evident at incident fluences considerably below the ablation threshold [Figs. 2(e) and 2(f)]. Estimates of the catastrophic self-focusing distance z_f for each of these substrates using the empirical formula of Marburger[16] are in rough agreement with the onset fluences observed. The 1 mm thick BK-7 sample produced distinct rings in the transmitted profile [Fig. 2(f)] at incident energies above 5.4 μJ (where $z_f=1$ mm predicts 5.8 μJ).[16] The 1 mm LiF sample produced results similar to the 1 mm thick borosilicate glass. Some flattening was evident above incident energies of 35 μJ (where $z_f=1$ mm predicts 32 μJ), but was accompanied by a ring-like intensity distribution. In the 1 mm thick sapphire sample, nonlinear optical effects led to excellent flattening of the transmitted beam [Fig. 2(d)] at incident energies just above $z_f=1$ mm (8 μJ both measured and calculated). However, the transmitted intensity was too low to be useful for shock generation. Higher incident fluences led to severe self-focusing and other bulk nonlinear optical effects [e.g., Fig. 2(e)]. Longer pulse lengths (210 and 400 fs) moved the range for flattening to somewhat higher fluences, as was expected because of the dependence of z_f on pulse length, but still inadequate for shock generation.

The 100 μm thick sapphire sample produced some flattening of the transmitted beam profile at fluences just above the ablation threshold, similar to the borosilicate cover glass. However, the flattening was not very uniform and there was a limited range of useful fluences. Also, the useful fluences were well above the ablation threshold in contrast to the borosilicate cover glass case, suggesting predominance of Kerr focusing similar to the effect seen in the 1 mm sapphire sample.

Future work to optimize the flatness, reproducibility, and transmitted beam fluence (to achieve stronger shocks) will involve varying the substrate material and thickness, as well as the incident laser pulse duration and profile.

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[1] D. L. Paisley, in *Shock Waves in Condensed Matter—1989*, edited by S. C. Schmidt and W. C. Tao (Elsevier Science, New York, 1990), pp. 733–736.
[2] W. M. Trott, in *High Pressure Science and Technology—1993*, edited by S. C. Schmidt, J. W. Shaner, G. A. Samara, and M. Ross (AIP, Woodbury, NY, 1994), pp. 1655–1658.
[3] R. Massudi and M. Piché, Opt. Commun. **142**, 61 (1997).
[4] F. M. Dickey and S. C. Holswade, Opt. Eng. **35**, 3285 (1996).
[5] B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, Phys. Rev. B **53**, 1749 (1996).
[6] E. E. B. Campbell, D. Ashkenasi, and A. Rosenfeld, Mater. Sci. Forum **301**, 123 (1999).
[7] M. Lenzner, J. Krüger, S. Sartania, Z. Cheng, Ch. Spielmann, G. Mourou, W. Kautek, and F. Krausz, Phys. Rev. Lett. **80**, 4076 (1998).

[8] A.-C. Tien, S. Backus, H. Kapteyn, M. Murnane, and G. Mourou, Phys. Rev. Lett. **82**, 3883 (1999).
[9] S. A. Diddams, H. K. Eaton, A. A. Zozulya, and T. S. Clement, Opt. Lett. **23**, 379 (1998).
[10] D. Ashkenasi, H. Varel, A. Rosenfeld, S. Henz, J. Herrmann, and E. E. B. Campbell, Appl. Phys. Lett. **72**, 1442 (1998).
[11] K. T. Gahagan, D. S. Moore, D. J. Funk, R. L. Rabie, S. J. Buelow, and J. W. Nicholson, Phys. Rev. Lett. **85**, 3205 (2000).
[12] J. P. Geindre, P. Audebert, A. Rousse, F. Falliès, J. C. Gauthier, A. Mysyrowicz, A. Dos Santos, G. Hamoniaux, and A. Antonetti, Opt. Lett. **19**, 1997 (1994).
[13] E. Tokunaga, A. Terasaki, and T. Kobayashi, Opt. Lett. **17**, 1131 (1992).
[14] C. Momma, S. Nolte, B. N. Chichkov, F. v. Alvensleben, and A. Tünnermann, Appl. Surf. Sci. **109**, 15 (1997).
[15] S. R. Greenfield (personal communication); E. Quéré, S. Guizard, P. Martin, G. Petite, O. Gobert, P. Meynadier, and M. Perdrix, Appl. Phys. B: Lasers Opt. **68**, 459 (1999).
[16] J. H. Marburger, Prog. Quantum Electron. **4**, 35 (1975).