Diode-pumped all-solid-state ultrafast Cr:LiSGAF laser oscillator–amplifier system applied to laser ablation


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Abstract

We describe a multi-kHz repetition rate ultrafast all-solid-state diode-pumped oscillator–regenerative amplifier laser system that we have applied to ultrafast laser ablation. The oscillator was a Kerr lens mode-locked Cr:LiSGAF laser and the regenerative amplifier utilised Cr:LiSGAF in a simple three-mirror cavity. The whole laser system, which was pumped by less than 2 W total pump power from four commercially available 670 nm diodes, produced ~1 μJ pulses at up to 10 kHz repetition rate, tunable in the near infrared. A simple double pass two grating compressor was used to adjust the pulse duration from 15 ps to 150 fs. Using this laser system we performed ablation of stainless steel and fused silica and demonstrated the characteristic pulse duration dependence of the ablation threshold for dielectrics. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The invention of chirped pulse amplification (CPA) [1] and the subsequent development of versatile ultrafast laser amplifier systems has led to many new areas of high intensity physics and to new applications of lasers. Ultrafast laser ablation is proving particularly rich in terms of scientific advances and potential applications. In this letter we demonstrate the application of a novel all-solid-state diode-pumped microjoule laser system, with its advantages of low cost and straightforward deployment, to ultrafast ablation of metals and dielectrics.

Due to their high peak intensities, ultrashort (picosecond and femtosecond) pulses ablate material via the rapid creation of a plasma via photoionization that absorbs the incident energy, resulting in direct vaporisation from the target surface (see e.g. Refs. [2,3]). This produces negligible collateral heating and shock-wave damage in comparison to conventional continuous wave and long pulse (nanosecond) laser ablation where the dominant process involved is the heating of the target material through the liquid phase to the vapour phase. In metals it becomes possible to drill holes and machine features with unprecedented precision, see e.g. Refs. [4,5]. In di-
electrics the plasma generation may be initiated by multi-photon ionisation rather than resonant absorption, resulting in an ablation threshold that, for sufficiently short pulses, becomes independent of local material properties, see e.g. Ref. [3]. This intensity dependence permits the ablation of features smaller than the focal spot size when operating near threshold, since only a fraction (the central region) of the focused light exceeds the threshold intensity [4].

This deterministic behaviour of ultrafast laser ablation provides great potential for application in surgery where microjoule level pulse energies are used and the margins for error in the ablation process are minimal, see e.g. Ref. [6]. A particularly exciting prospect is the LASIX procedure to reshape the cornea. Other real-world applications include the manufacture of microprosthetics, the drilling of precision holes in ink jet nozzles and fuel injection systems and the microdissection of biological tissue in novel diagnostic and analytic instrumentation. Most of the ablation work to date, however, has been carried out using expensive systems built around large frame lasers that are difficult to implement in situations like a surgical theatre or a biomedical instrument. Instead they require specialist laser environments with appropriate water cooling, electricity and maintenance provisions. It is thus important to demonstrate that these potential applications of ultrafast ablation can be realised using relatively low cost and potentially compact laser technology.

The laser system reported here is similar to the first such all-solid state diode-pumped oscillator–amplifier system reported in Ref. [7] except that it has been designed to exploit Cr:LiSGAF, rather than Cr:LiSAF, as the gain medium and is constructed from components that are all commercially available, making the industrial manufacture of such a system entirely feasible. It has been reported that Cr:LiSGAF provides superior performance to Cr:LiSAF in Q-switched lasers [8] and our new laser system is considerably more straightforward to align and maintain at a useful level of performance, providing tunable microjoule energy pulses at kilohertz repetition rates for a total of three 500 mW and one 350 mW pump diodes.

2. Cr:LiSGAF Kerr lens mode-locked oscillator

The Kerr lens mode-locked femtosecond oscillator, shown in Fig. 1, was based on an asymmetric four-mirror cavity design [9], incorporating one 50

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**Fig. 1.** Diode-pumped Kerr lens mode-locked femtosecond oscillator. L1: spherical lens ($f = 4.5$ mm); L2: cylindrical lens ($f = 6.4$ mm); L3: cylindrical lens ($f = 100$ mm); L4: spherical lens ($f = 50$ mm); L5: cylindrical lens ($f = 200$ mm); L6: spherical lens ($f = 100$ mm); M1 and M2: folding mirrors (ROC = 50 and 100 mm, respectively); M3: 0.7% output coupler; M4: high reflector; P1: intracavity prism; P2: intracavity prism with AOM; S1: mode-locking aperture; S2: tuning slit; and PD: photodiode.
Fig. 2. Average output power as a function of wavelength of Cr:LiSGAF oscillator for CW (upper curve, circular symbols) and mode-locked operation with: two pump diodes (middle curve, triangular symbols) and one pump diode (bottom curve, square symbols).

mm radius of curvature (ROC) folding mirror and one 100 mm ROC folding mirror. The 6 mm long Brewster/Brewster-cut Cr:LiSGAF laser rod (3% Cr$^{3+}$) was longitudinally pumped by one 350 mW, 50 μm stripe diode (LD1) and one 500 mW, 100 μm stripe diode (LD2), both emitting at 670 nm. Each diode beam was first collimated by a high numerical aperture aspherical lens, expanded by a cylindrical telescope in the horizontal axis and focused through the folding mirrors into the rod by a spherical lens.

Two intracavity quartz prisms were used for dispersion compensation (separation: 80 cm) in one arm of the oscillator. The mode-locking was regeneratively initiated using an acousto-optic modulator incorporated into one of the intracavity prisms. This was electronically driven at the cavity round trip time (∼80 MHz) by a unit (Kentech Instruments) which was triggered by the optical signal leaking through the high reflector (M4). Kerr lens mode-locked operation was thus initiated and sustained indefinitely.

Fig. 2 shows the tuning characteristics of the oscillator in CW operation with two pump diodes and Kerr lens mode-locked with both one diode and two pump diodes. Although the tunability of the laser is not critical for the application of ultrafast ablation, it is useful for other applications, including fluorescence lifetime imaging [10] and two photon microscopy. Fig. 3 shows a typical autocorrelation trace and corresponding spectrum of the output of the oscillator for an 80 MHz pulse train of 100 fs pulses. The maximum average output power was 31 mW when pumping with both diodes and 15 mW when pumping with 250 mW from the 50 μm stripe diode, using a 0.7% output coupler.

3. Cr:LiSGAF regenerative amplifier

The 1 m long regenerative amplifier cavity was a three-mirror design incorporating a mirror coated on the plane face of a 5 mm long plane/Brewster-cut Cr:LiSGAF rod (3% Cr$^{3+}$) with a 100 mm ROC folding mirror and a plane high reflector. Fig. 4 shows a schematic of the amplifier. Cr:LiSGAF was
chosen as the gain medium over Cr:LiSAF due to its reduced thermal quenching problems and longer upper state lifetime (~ 84 μs). The rod was pumped through both faces by two 500 mW 100 μm stripe diodes at 670 nm. Each diode beam was shaped in a similar manner to the diode pump beams for the oscillator.

With a 0.5% output coupler in place of M3, this arrangement produced 270 mW of CW power and a sub-mW laser threshold pump power. For Q-switched operation and regenerative amplification, this output coupler was replaced with a high reflector. A broadband thin film polariser (Alpine Research Optics) and Pockels cell (Medox Electro-Optics) were used to Q-switch the cavity by means of a quarter wave polarisation rotation induced by the Pockels cell. The shortest Q-switch time achieved (the time between allowing cavity to lase and the actual peak of the Q-switch pulse) was ~ 1.4 μs. When the amplifier cavity was seeded with 10 pJ, 100 fs pulses from the

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**Fig. 4.** Diode-pumped regenerative amplifier: L1: spherical lens (f = 4.5 mm); L2: cylindrical lens (f = 6.4 mm); L3: cylindrical lens (f = 100 mm); L4: spherical lens (f = 50 mm); L5: cylindrical lens (f = 200 mm); L6: spherical lens (f = 80 mm); M2: folding mirror (ROC = 100 mm); M3: high reflector; and TFP: thin film polariser.

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**Fig. 5.** Autocorrelation trace (a) and corresponding spectrum (b) of output of amplifier for 13 ps pulses assuming sech² pulse shape (Δλ = 11.5 nm).
The amplified pulses reached their maximum energy of \( \sim 1 \, \mu J \) in as little as \( \sim 1.2 \, \mu s \). The dispersion of the regenerative amplifier cavity resulted in an amplified pulse duration of typically 13 ps, assuming a sech\(^2\) pulse shape.

One of the potential difficulties associated with pulse amplification is the accumulative effect of self-phase modulation, due to the high peak intensities present in the cavity. This can distort the pulse spectrum and prevent effective recompression. We did not observe such spectral distortions (see Fig. 5) and presume that the group velocity dispersion (GVD) in the regenerative amplifier cavity kept the peak pulse power low enough to avoid this problem. We therefore did not need to employ a pulse stretcher prior to seeding the amplifier and this made the system much more straightforward to set up and to align. Fig. 5 shows a typical autocorrelation trace with corresponding spectrum of the dispersed output pulses from the amplifier.

A simple double pass two grating compressor was used to provide extracavity negative GVD and compress the pulses down to 1.5 times their duration (100 fs) from the oscillator. We assume that further compression was limited by the accumulation of third order dispersion contributions in the amplifier cavity and grating compressor. We note that there are now many established approaches to compensating for higher order dispersion contributions in ultrafast pulse amplification schemes and these could be applied in this case.

The spectral performance of the amplifier was investigated by tuning the oscillator across its full range and measuring the corresponding amplified pulse energy. These results can be seen in Fig. 6 where they show only a 10% variation in pulse energy between 820 nm and 855 nm.

The repetition rate of the amplifier was varied from 150 Hz to 20 kHz and this demonstrated less than a 10% decrease in pulse energy up to 10 kHz, as illustrated in Fig. 7. Above 10 kHz the pulse energy falls away since the repetition period approaches the upper-state lifetime of the Cr:LiSGAF crystal (84 \( \mu s \)). The output beam of the amplifier had a measured \( M^2 \) value of 1.19 uncompressed and a measured \( M^2 \) value of 1.73 after the grating compressor. This degradation was attributed to the poor condition of the diffraction gratings used.

### 4. Ultrafast laser ablation experiments

To confirm that this laser system can indeed be applied to ultrafast laser ablation, a study was made of the ablation of stainless steel and fused silica as a function of pulse duration. By varying the grating separation in the compressor the pulse duration \( \tau \) at the target was varied from 13 ps to \( \sim 150 \, \text{fs} \).

The output from the amplifier was directed through the double grating compressor to the experimental ablation set-up, which allowed the routine switching between target and autocorrelator and included a permanent beam pick-off (\( \sim 4\% \)) directed
onto a calibrated monitor photodiode. In this manner it was possible to perform ablation of a sample and measure the corresponding pulse energy and duration at the same time. The positioning of the target in the plane of its surface (x–y plane) was achieved manually using two micrometer translation stages and the positioning in the axis perpendicular to the surface (z-axis) was controlled electronically by a sub-micron positioning actuator. The pulses were focused onto the target using a 4.5 mm focal length aspherical lens, producing a focused spot estimated to be 0.9 μm radius when using the uncompressed picosecond pulses and 1.4 μm radius when using the compressed pulses. In all cases, each hole was ablated with ~320 pulses as the target was scanned at constant velocity in the z-axis through the focus of the beam whilst the amplifier operated at 5 kHz.

A 5 mW He–Ne laser beam (with a rotating diffuser) was used to illuminate the sample surface, which was imaged onto a CCD camera to provide in situ observation (with a resolution of ~1 μm) of the sample surface during irradiation.

Fig. 8 shows electron micrograph images of holes ablated in a polished stainless steel target (note the different scales). The holes shown in the two top images were ablated using 10 ps pulses and the holes shown in the two bottom images were ablated using 150 fs pulses. The holes are approximately 2 μm in diameter in both cases and the features are very similar, suggesting that for many applications, one...
can do without the compressor and so reduce the complexity and cost of the system. We did, however, observe a modest improvement in uniformity and repeatability for the femtosecond pulses. Since a vacuum chamber was not utilised and the target was not cleaned before inspection, the material that was ejected from the hole settled around the rim and can still be seen.

Fig. 9 shows CCD camera images of holes ablated in fused silica as a function of decreasing pulse energy. In order to demonstrate the differences in the ablation process for different pulse durations more clearly, we investigated the ablation threshold as a function of pulse duration for both materials. The ablation threshold was determined by monitoring the sample surface (as described above) and observing the onset of ablation as evinced by the appearance of the hole and associated debris. While this is not an adequate method for determining absolute ablation thresholds, it proved sufficient to demonstrate relative changes in threshold behaviour.

Fig. 10 shows the dependence of the ablation threshold fluence on pulse duration for both stainless steel and fused silica. In the case of the dielectric sample the characteristic deviation from the square-root dependence of the ablation threshold on the pulse duration can be seen for ultrashort pulses. This deviation can be understood in terms of multi-photon absorption initiating the avalanche breakdown of the material [2,3]. For the metal sample there was no significant change in the ablation threshold energy over our pulse duration range as is expected as the ablation process is different from that of a dielectric due to the presence of free charge carriers [11].

![Fig. 9. Diode-pumped ultrafast ablation of fused silica ablated with 140 fs pulses (top 4 holes) and 5 ps pulses (bottom 4 holes).](image)

![Fig. 10. Ablation threshold fluence of stainless steel and fused silica for varying pulse durations.](image)
These results demonstrate some of the basic features of the ultrafast ablation process using this microjoule level pulse energy laser system.

5. Conclusions

We have demonstrated a versatile ultrafast all-solid-state diode-pumped oscillator–amplifier system, based on Cr:LiSGAF for the first time, which provides microjoule energy level pulses whose energy, wavelength, pulse repetition rate and duration may be adjusted to suit a variety of applications. This laser system incorporates just four commercially available pump diodes requiring less than 4 amperes total current, including cooling. This makes it potentially low cost and widely deployable. Higher pulse energies may be achieved by incorporating two additional pump diodes using polarisation multiplexing. Still further pulse energies could be realised at 10 kHz repetition rates by employing high power diode bars to pump a thin Cr:LiSGAF slab as described in Ref. [12].

We have applied this laser system to the ablation of both metal and glass with varying pulse durations and sub-μJ ablation thresholds, thereby demonstrating its suitability to high precision micromachining applications, including surgery. We note that for precise ablation, the requirement to minimise processing time while operating just above ablation threshold requires a high repetition rate laser system. The maximum desired repetition rate is fixed by the thermal response time of the target and 10 kHz appears to be close to optimum for most applications. This repetition rate is straightforward to achieve in Cr:LiSGAF but rather challenging in diode-pumped Nd laser based systems including Ti:sapphire amplifiers.

Beyond micromachining, there are many other scientific and technological applications for ultrafast microjoule class laser systems. We have applied this technology to fluorescence lifetime imaging [10] and all-optical storage based on reversible amorphous-crystalline phase changes [13]. Three-dimensional optical storage with sub-μm resolution [14] would provide another interesting application.

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