1. Ultrashort-Pulse Generation and Ultrafast Phenomena

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1.1 Novel Mode-Locked Laser Cavity Designs for High Peak Intensities

Passive modelocking of ultrashort pulsed lasers has been an active research area in recent years, resulting in record pulse durations and simpler, more compact laser designs. This technology has been applied to biomedical optics, high speed communications, and the investigation of ultrafast nonlinear processes in semiconductor materials and devices. In order to be useful for widespread commercial applications, ultrashort pulse laser sources must be technologically simple, robust, and cost effective. Kerr-lens modelocking (KLM) of solid state lasers has been the most successful approach for short pulse generation. KLM is a passive modelocking technique which utilizes the Kerr effect, or nonlinear index of refraction, to create an artificial fast saturable absorber. This technique combined with intracavity dispersion management can yield extremely short pulse durations. Working in collaboration with Professors Erich P. Ippen and Hermann A. Haus, we have recently demonstrated the generation of pulses of ~5 fs duration from a $Ti:Al_2O_3$ laser. These pulses are less than two optical cycles and are the shortest ever produced directly from a laser oscillator.[1] In addition to experimental studies, we have also developed theoretical models which provide a foundation for understanding and optimizing short-pulse KLM lasers.[2,3] Our program investigates several areas of ultrafast laser technology, with the objective of developing new technologies that can be applied across a range of laser materials and systems.

An important goal in ultrafast optics is to increase laser output pulse energies and intensities. Since the total output power of the laser generally cannot be increased, the pulse energies can be increased by increasing cavity lengths and therefore reducing the laser repetition rate. Other investigators have also examined techniques for designing low repetition rate KLM lasers that

[1] U. Morgner, F.X. Kärtner, S.H. Cho, Y. Chen, H.A. Haus, J.G. Fujimoto, E.P. Ippen, V. Scheurer, G. Angelow, and T. Tschudi, "Sub-Two Cycle fs-Pulses from a Kerr-Lens Modelocked Ti:sapphire Laser," *Opt. Lett.,* 24: 411-413 (1999).

[2] H.A. Haus, J.G. Fujimoto, and E.P. Ippen, "Structures for Additive Pulse Modelocking," *J. Opt. Soc. Am. B* 8:2068 (1991); H. A. Haus, J. G. Fujimoto, and E. P. Ippen, "Analytic Theory of Additive Pulse and Kerr Lens Mode Locking," *IEEE J. Quant. Electron.* 28: 2086 (1992).

[3] Y. Chen, F.X. Kärtner, U. Morgner, S.H. Cho, H.A. Haus, E.P. Ippen, and J.G. Fujimoto, "Dispersion Managed Mode-Locking," *JOSA B* 16: 1999-2004 (1999).

produce high-intensity pulses.[4,5] Typical short pulse lasers have cavity lengths of approximately 1 meter and operate with 100 MHz repetition rates. Designing long cavity lasers enables the repetition rate to be reduced to as low as a few MHz. This reduced repetition rate is a significant advantage for nonlinear and ultrafast studies, because it reduces thermal parasitics, sample damage problems, and recovery time artifacts. Also, increasing pulse energy without using cavity dumpers or amplifiers yields a low cost and simple system. Generation of femtosecond (fs) pulses with high intensities in the MW range is essential for a number of applications including optical harmonic generation and investigation of ultrafast nonlinear optical phenomena. The peak power directly generated by typical modelocked Ti:Al_2O_3 laser sources is in the range of hundreds of kW which is often insufficient for studies of nonlinear phenomena. The development of low cost laser sources with high pulse energies will enable a wider range of femtosecond measurement applications, making this technology more available to both the research and the development communities.

The cavity length of our laser is extended by use of a Herriott-style multi-pass cavity (MPC), which is used for optical delay lines. This method has the advantage of providing a long path length but having a unity q parameter transformation.[6] This allows the cavity length to be increased while keeping the KLM operating point nearly invariant. Thus, if this device is inserted into the KLM laser, it has a zero effective length and leaves the laser cavity mode and nonlinear focusing behavior invariant. The development of long cavity fs-lasers requires careful design because the laser cavity must be operated in a particular subset of its stability region for optimum mode locking performance.

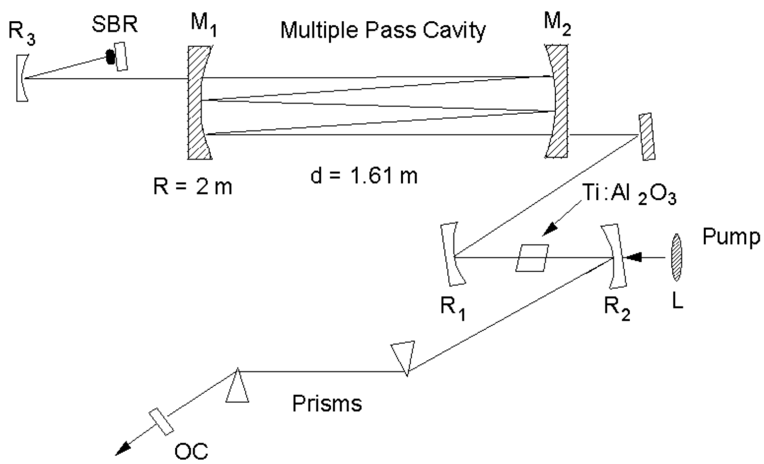

Figure 1: Ti:Al_2O_3 laser schematic with a pair of multiple pass cavity mirrors: M1, M2, 2" diameter MPC mirrors(Spectra-Physics) with R = 2 m radius of curvature; R1, R2, 10 cm radius of curvature mirrors (Spectra-physics); L, 6.3 cm focal length pump beam focusing lens;OC, 27% ½" thick output coupler (Spectra-Physics); Prisms, fused silica prisms.

Following this approach, we designed a high peak power laser using a standard, dispersion compensated KLM Ti:Al_2O_3 resonator with an MPC incorporated into one of the arms (Figure 1).

[4] A.R. Libertun, R. Shelton, H.C. Kapteyn, M.M. Murnane, "A 36 nJ-15.5 MHz Extended-Cavity Ti:Sapphire oscillator," paper presented at the 1999 Conference on Lasers and Electro-Optics, Baltimore, Maryland, May 22-28, 1999.

[5] A. Poppe, M. Lenzner, F. Krausz, and Ch. Spielmann, " A Sub-10 fs, 2.5 MW Ti:Sapphire oscillator," paper presented at the 1999 conference on Ultrafast Optics, Zurich, Switzerland, 1999.

[6] D. Herriott, H. Kogelnik, and R. Kompfner, "Off-Axis Paths in Spherical Mirror Interferometers," *Appl. Opt.* 3: 523-26 (1964); B. Perry. R.O.Brickman, A. Stein, E. B. Treacy, and P. Rabinowitz, " Controllable Pulse Compression in a Multiple-pass Raman Laser," *Opt. Lett.* 5: 288 (1980).

Operation at 15 MHz repetition rate was achieved using a design in which the beam made 20 round trips between the MPC mirrors separated by 82.4 cm.[7,8] Stable Kerr-lens modelocking was achieved resulting in 0.7 MW peak power and 16.5 fs nearly transform limited pulses.[9] Lower repetition rates of 7.2 MHz were achieved using 24 round trips between the MPC mirrors separated by 148 cm. We obtained 21 nJ output pulse energy in the two output beams and 23.5 fs nearly transform limited pulses, with a peak power of 0.9 MW. At a 5 MHz repetition rate, the beam makes 32 round trip passes between the MPC mirrors separated by 161 cm (Figure 2). Stable 60 nJ pulse energies with 44 fs nearly transform-limited pulses and a peak power of 1.4 MW were obtained (Figure 3a).

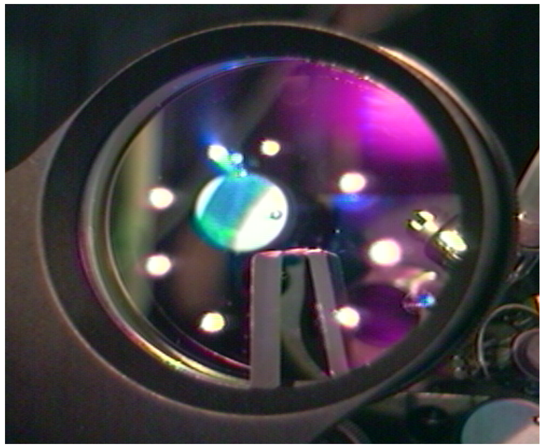

Figure 2: Photograph of an MPC mirror with 8 bounces per mirror used in a 5 MHz Ti:Al_2O_3 laser with a separation of 161 cm between two MPC mirrors.

However, it is difficult to reduce pulse repetition rates because the peak pulse intensity increases, producing multiple pulsing instabilities. This necessitates laser operation in a regime with lower self-amplitude modulation KLM nonlinearity, making the mode locking difficult to start. Thus, the starting mechanism for KLM should be decoupled from steady state mode-locked operation. To assist starting and to stabilize the laser against multiple pulsing, a saturable Bragg reflector (SBR)[10] is incorporated in the 7.2 MHz and 5 MHz lasers. Since it is hard to start mode locking in the single pulsing regime as the total cavity length is long, the use of the SBR was essential to obtaining stable mode locking operation with these long cavity lengths. The structure of the SBR consists of an AlAs/$Al_{0.85}Ga_{0.15}As$ quarter-wave dielectric stack and a single GaAs quantum well grown by molecular beam epitaxy. These studies are being performed in collaboration with Dr. Wayne Knox from Lucent Technologies.

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[7] S.H. Cho, B.E. Bouma, E.P. Ippen, and J.G. Fujimoto, "A 15 MHz, 0.5 MW KLM Ti:Al_2O_3 Laser using Multiple Pass Cavity," paper presented at the 1998 Conference on Lasers and Electro-Optics, San Francisco, California, May 2-8, 1998.

[8] S.H. Cho, B. E. Bouma, E.P. Ippen, and J.G. Fujimoto, "A Low Repetition Rate High Peak Power KLM Ti:Al_2O_3 Laser with a Multiple Pass Cavity," *Opt. Lett.* 24: 417-419 (1999).

[9] S.H. Cho, U. Morgner, F.X. Kärtner, E. P. Ippen, J.G. Fujimoto, J.E. Cunningham, and W.H. Knox, "A 7.2 MHz High Power KLM Ti:Al_2O_3 Laser using a Multiple Pass Cavity and Saturable Bragg Reflector," paper presented at the 1999 Conference on Lasers and Electro-Optics, Baltimore, MD, May 22-28, 1999.

[10] S. Tsuda, W.H. Knox, S.T. Cundiff, W.Y. Jan, and J.E. Cunningham, "Mode-Locking of Ultrafast Solid-State Lasers with Saturable Bragg Reflectors," *IEEE J. Select. Top. Quantum Electron*. 2: 454-64 (1996).

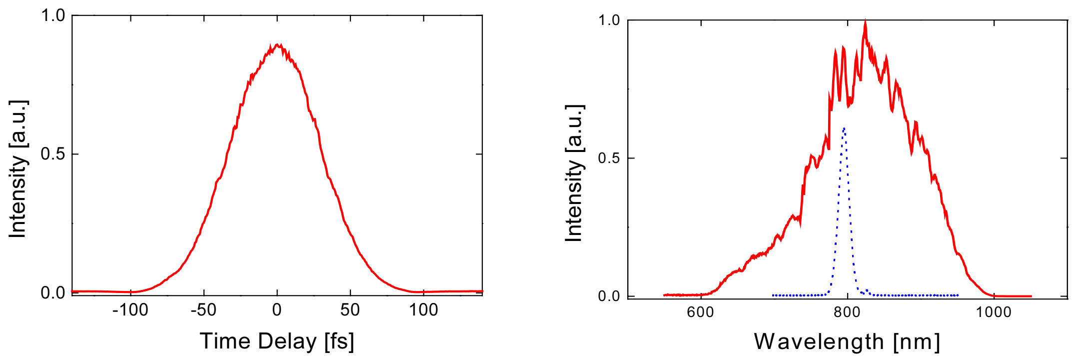

Figure 3: (a) Intensity autocorrelation trace showing a pulse duration of 44 fs assuming a $sech^2(t)$ intensity profile, (b) Continuum generation through a 0.55 NA focusing lens into a single mode fiber (3M, FS-SC-3314); spectrum of an initial pulse (dotted line), generated continuum (solid line).

This high pulse energy has been applied to generate continuum by focusing into a short length of single-mode optical fiber. The broadened spectrum of the pulses after passing through the fiber extends from 600 nm to 1000 nm (Figure 3b). Spectral broadening or continuum generation could also be produced by tightly focusing the output beam into the transparent material in which the peak power is approximately above the threshold for self-focusing. The high pulse energies and repetition rates from this laser also enable applications to three dimensional optical memory and micromachining or microsctructuring using nonlinear material interaction with fs-pulses.

1.2 New Ultrashort Pulse Laser Sources Using Double-Chirped Mirrors for Dispersion Compensation

The generation of extremely short pulse durations is important for a variety of applications since the pulse durations determine the highest resolution ultrafast measurements that can be performed. The broad bandwidths associated with extremely short pulses also enable spectrally resolved measurements or the generation of short pulses at other wavelengths. Pulses as short as 4 to 5 fs have been produced by external compression of high energy amplified femtosecond pulses. However, the laser sources producing those high-energy pulses at 1 kHz[11] and 1 MHz[12] are relatively complex and expensive. For applications such as ultrahigh-resolution time domain spectroscopy or optical coherence tomography (OCT), high performance ultrashort pulse sources are required. In this project, pulses shorter than 5.4 fs at a center wavelength of 800 nm, corresponding to a bandwidth greater than 350 nm, have been generated directly by a Kerr-lens mode-locked Ti: Al_2O_3 laser at a repetition rate of 90 MHz and an average output power of 200 mW. This world-record pulse duration is achieved by compensating the high order intracavity dispersion using specially-designed and fabricated double-chirped mirrors (DCMs). These studies have been performed in collaboration with Professors Erich Ippen, Hermann Haus, and Franz Kaertner.

In chirped mirrors, variation of the Bragg wavelength during the layer growth leads to deeper penetration of longer wavelengths into the mirror structure.[13] This results in a broad reflectivity

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[11] M. Nisoli, S. de Silvestri, O. Svelto, R. Szipocs, K. Ferencz, C. Spielmann, S. Sartania, and F. Krausz, "Compression of high-energy laser pulses below 5 fs," *Optics Letters* 22(8): 522-4 (1997).

[12] A. Baltuska, W. Zhiyi, M. S. Pshenichnikov, and D. A. Wiersma, "Optical pulse compression to 5 fs at a 1-MHz repetition rate," *Optics Letters* 22(2): 102-4 (1997).

[13] R. Szipöcs, K. Ferencz, C. Spielmann, and F. Krausz, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," *Optics Letters* 19(3): 201-3 (1994).

range and a negative dispersion of the reflected pulse. In DCMs, the coupling coefficient of the Bragg mirror is also chirped by gradually increasing the thickness of the high index layer; this minimizes oscillations in the group delay dispersion and enables modelocking over the full high reflectivity range of the mirror. Since the phase characteristics of the 54 layers are extremely sensitive to variations in the layer thickness, our DCMs were grown by ion-beam sputtering, the state-of-the-art technique for dielectric multi-layer depositing of DCMs.[14] Mirror fabrication was performed by investigators at the University of Darmstadt, Germany.

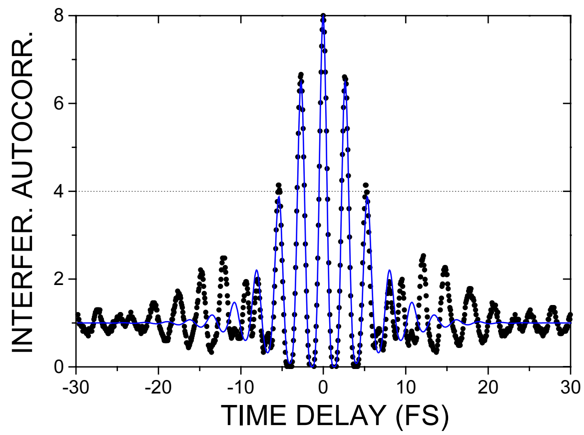

Figure 4: Measured interferometric autocorrelation (dots) of the Ti:Al_2O_3 laser pulses. The dashed curve is a fit of $sech^2$-function with a FWHM of 5.4 fs, and the solid curve is the calculation from the spectrum.

The Ti: Al_2O_3 laser uses a standard z-fold cavity-design. The Ti:Al_2O_3 crystal (thickness 2.05 mm) is placed between two curved mirrors with 100-mm radius of curvature. All mirrors in the cavity are DCMs, except the output-coupling mirror and one silver mirror. An interferometric autocorrelation of the ultrashort pulses is shown in Figure 4 (dots); the autocorrelator is a standard interferometric design[15] with two counteroriented metallic beam beamsplitters on a 0.1-mm substrate and a 30-µm thin KDP crystal. A fit to a sech-shaped pulse would result in a pulse width of 4.3 fs FWHM, a fit to a Gauss-shaped pulse in 4.8 fs. The most conservative assumption of a sinc-shaped pulse gives a pulse duration of 5.4 fs (dashed curve). The good agreement of the measured autocorrelation with the autocorrelation function derived from the spectrum (solid line) indicates a pulse width between 4.9-5.4 fs, which corresponds to less than two optical cycles at the center wavelength of 800 nm. Sub-two-cycle pulses might be used to study novel nonlinear optical effects that depend on the phase between carrier and the envelope.[16]

Another interesting wavelength range is centered around 1300 nm, in which falls the regime of zero group-velocity dispersion of most materials. In addition, broadband lasers at 1.3 µm are important for biomedical optical imaging and optical coherence tomography because they have increased penetration depth in tissue.[17,18] We have designed a high performance Cr:forsterite

[14] M. Tilsch, V. Scheurer, J. Staub, and T. Tschudi, 1994 (SPIE), p. 414.

[15] C. Spielmann, X. Lin, and F. Krausz, "Measurement of Interferometric Autocorrelations: Comment," *Applied Optics* 36(12): 2523-5 (1997).

[16] F. Krausz, T. Brabec, M. Schnürer, and C. Spielmann, "Extreme Nonlinear Optics: Exposing Matter to a Few Periods of Light," *Optics & Photonics News* (July): 46-51 (1998).

[17] B.E. Bouma, M. Ramaswamy-Paye, and J.G. Fujimoto, "In Vivo Endoscopic Optical Biopsy with Optical Coherence Tomography," *Science* 276(5321): 2037-9 (1997).

laser using custom DCMs for dispersion compensation. The cavity is a standard z-fold design using a 5-mm Brewster-cut Cr:forsterite crystal pumped by a diode-pumped Nd:YAG laser. All mirrors except the output-coupling mirror are DCMs.

The dispersion characteristics of all material inside the cavity determine the phase characteristics of the DCMs. Because low-loss mirrors are needed for the low-gain material Cr:forsterite, the amplitude characteristics of the DCMs are determined by the criterion of a 99.8% reflectivity over as much bandwidth as possible. At the same time, maximum transmission is maintained at the pump wavelength of 1.064 μm. These DCMs were also grown by the ion-beam sputtering technique.

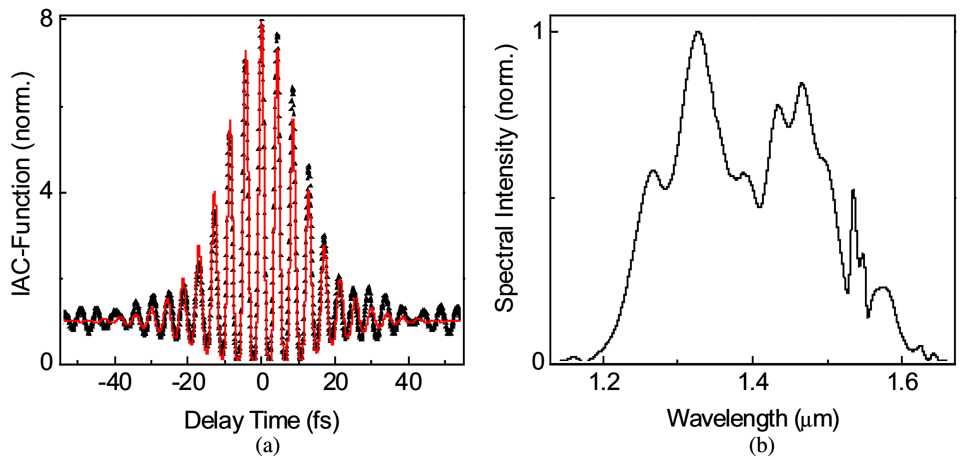

Figure 5: (a) Measured autocorrelation of the Cr:Forsterite laser output showing 14 fs pulse duration. (b) Measured laser output spectrum. These pulse durations are made possible using intracavity dispersion management with double chirped mirrors (DCM's). The pulse duration is the shortest ever generated from a Cr:Forsterite laser.

Using dispersion compensation with DCMs, we have generated pulses with durations as short as 14 fs. These are the shortest pulse durations generated to date from a Cr:forsterite laser. The laser spectrum has a bandwidth (FWHM) of 250 nm and extends from 1200-1470 nm as shown in Figure 5. Previous investigators have demonstrated pulses as short as 20 fs and bandwidths of 90 nm produced by a Kerr-lens mode-locked Cr:forsterite laser.[19] Our results are still preliminary, and we expect that shorter pulses can be obtained with further optimization.

1.3 Development of Nonepitaxially Grown Saturable Absorber Materials for Laser Modelocking

In recent years many ultrafast solid state lasers have used semiconductor saturable absorber devices for short pulse generation.[20] KLM is a common method for ultrashort pulse generation; however, KLM typically requires careful optimization of the laser cavity and operating point. In

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[18] B.E. Bouma, G.J. Tearney, S.A. Boppart, B. Golubovic, I.P. Bilinsky, M.E. Brezinski, and J.G. Fujimoto, "Mode Locked Solid State Laser Sources for Optical Coherence Tomography," *SPIE Int. Soc. Opt. Eng. Proceedings of Spie the International Society for Optical Engineering* 2981: 37-44 (1997).

[19] Z. Zhang, K. Torizuka, T. Itatani, K. Kobayashi, T. Sugaya, and T. Nakagawa, "Femtosecond Cr:forsterite Laser with Mode Locking Initiated by a Quantum-Well Saturable Absorber," *IEEE Journal of Quantum Electronics* 33(11): 1975-81 (1997).

[20] U. Keller, D.A.B. Miller, G.D. Boyd, T.H. Chiu, J.F. Ferguson, and M.T. Asom, "Solid-State Low-Loss Intracavity Saturable Absorber for Nd:YLF lasers: An Antiresonant Semiconductor Fabry-Perot Saturable Absorber", *Opt. Lett*. 17: 505-507 (1992).

addition, KLM short pulse generation is usually not self-starting and is sensitive to external perturbations. Semiconductor saturable absorber devices provide several advantages, including self-starting operation, simplicity in laser cavity design, and decoupling of the gain and modelocking mechanisms.

The most common saturable absorber technologies are semiconductor saturable absorber mirrors[22] and saturable Bragg reflectors[23], which have been extensively used for both saturable absorber modelocking and in combination with KLM. However, these devices require epitaxial growth, which imposes lattice-matching constraints on the absorber materials and also requires complex and expensive systems for sample fabrication.

A promising alternative to epitaxially grown saturable absorbers is the use of non-epitaxially grown semiconductor doped silica films as saturable absorber devices. These devices are fabricated by doping semiconductor nanocrystallites into silica films using rf or magnetron rf sputtering.[24] RF sputtering is a simple and inexpensive technique that employs an argon plasma to eject material from a target which is then deposited onto a substrate. Magnetron rf sputtering improves deposition rates by confining the plasma to the target area.

Semiconductor doped silica films grown using rf or magnetron rf sputtering have several desirable features. They can be deposited on virtually any substrate, including oxides such as glass and dielectric coatings as well as metal mirrors. By varying the doping density of the semiconductor quantum dots, one can adjust the absorption coefficient of the device. A wide range of semiconductor materials can be doped into the silica films. Finally, appropriate choice of the semiconductor and knowledge of quantum confinement effects allow one to control the operating wavelength and absorption edge of the device.

In our previous work[25], InAs-doped silica films were successfully applied to laser modelocking. These films were fabricated using rf sputtering in collaboration with Dr. James N. Walpole and Leo J. Misaggia at Lincoln Laboratories and subjected to a rapid thermal annealing treatment; their structural and optical properties were then comprehensively characterized. With a InAs-doped silica film saturable absorber used in a standard z-cavity Ti:Al_2O_3 laser, self-starting saturable absorber assisted KLM was obtained, with pulses as short as 25 fs and a FWHM bandwidth of 53 nm. The laser wavelength was tunable from 800 to 880 nm while sustaining self-starting operation. The main difficulty with these devices was their high saturation fluence, making modelocking impossible without the aid of KLM.

Recently, we have investigated different approaches to reducing the saturation fluence of our nonepitaxially grown saturable absorbers. One approach was to use a magnetron rf sputtering system in collaboration with Professor Michael F. Ruane and Paul Mak at the Boston University Photonics Center to deposit GaSb-doped silica films on sapphire substrates. In general, larger quantum dots should lead to lower saturation fluences since the excitation is closer to the

[22] U. Keller, K.J. Weingarten, F.X. Kärtner, D. Kopf, B. Braun, I.D. Jung, R. Fluck, C. Hönninger, N. Matuschek, and J. Aus der Au, "Semiconductor Saturable Absorber Mirrors (SESAM's) for Femtosecond to Nanosecond Pulse Generation in Solid-State Lasers*", IEEE J. Select. Top. Quantum Electron.* 2: 435-53 (1996).

[23] S. Tsuda, W.H. Knox, S.T. Cundiff, W.Y. Jan, and J.E. Cunningham, "Mode-Locking of Ultrafast Solid-State Lasers with Saturable Bragg Reflectors", *IEEE J. Select. Top. Quantum Electron.* 2: 454-64 (1996).

[24] K. Tsunetomo, H. Nasu, H. Kitayama, A. Kawabuchi, Y. Osaka, and K. Takiyama, "Quantum Size Effect of Semiconductor Microcrystallites Doped in SiO_2-Glass Thin Flims Prepared by RF-Sputtering", *Jap. J. Appl. Phys.* 28: 1928-1933 (1989).

[25] I.P. Bilinsky, J.G. Fujimoto, L.J. Misaggia, and J.N. Walpole, "Semiconductor-Doped-Silica Saturable Absorber Films for Solid-State Laser Modelocking", *Opt. Lett.* 23(22): 1766-1768 (1998).

bandedge and there is less scattering on interface defects.[26] The magnetron rf sputtering system allows control over the substrate temperature, which has been shown to increase quantum dot sizes.[27] Using GaSb was also expected to lower the saturation fluence since its bulk bandgap is larger, resulting in larger quantum dots and excitation closer to the bandedge.

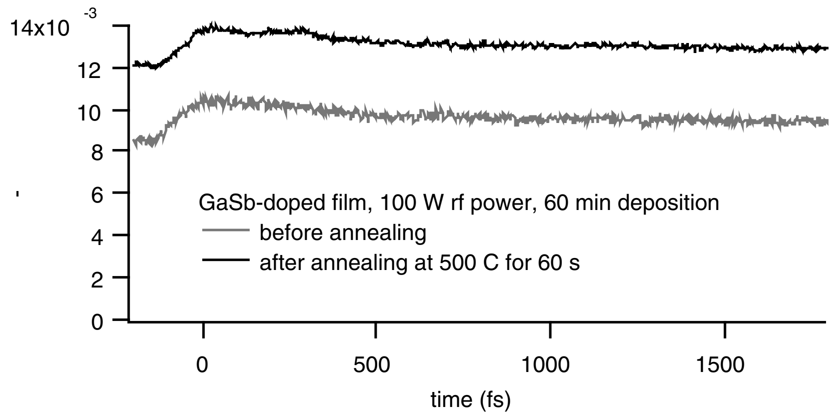

Figure 6: GaSb-doped silica films grown at 100 W for 60 min before and after rapid thermal annealing at 500°C for 60 seconds.

We measured the linear transmission of the GaSb-doped silica films and found that the transmission characteristics did not vary significantly as a function of either rf power or film thickness. Pump probe experiments were done on both annealed and unannealed films to characterize their nonlinear optical properties (Figure 6). As expected, unannealed GaSb-doped silica films had a lower saturation fluence than the unannealed InAs-doped silica films grown previously in a non-magnetron rf sputtering system. However, after annealing, the optical quality of the GaSb-doped silica films was considerably reduced and the saturation fluence increased, making these devices unsuitable for laser modelocking. To confirm this, we introduced these devices into a $Ti:Al_2O_3$ laser cavity, where no self-starting modelocking was observed. These studies showed that GaSb-doped silica films did not function as effective saturable absorbers.

We also examined the effects of varying the substrate temperature during growth on the GaSb-doped silica films. Pump probe experiments revealed strong oscillations in the signal which can be attributed to coherent phonon generation (Figure 7). The amplitude of these oscillations increased with substrate temperature. A simple theoretical model indicated that the oscillations were likely caused by optical phonons. Further investigations are needed to understand the mechanisms and possible applications of the high frequency reflectivity oscillations.

Another approach to reducing the saturation fluence was to incorporate semiconductor-doped silica films into a reflective structure. A standing wave is formed in a laser cavity when an electromagnetic wave is reflected from a mirror; by placing the semiconductor-doped film at the peak of the standing wave, the effective saturation fluence can be reduced by a factor of four.

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[26] I.P. Bilinsky, *Novel Saturable Absorber Materials and Devices for Laser Modelocking*, Ph. D. Thesis, Department of Physics, MIT, 1999.

[27] K. Tsunetomo, M. Yamamoto, and Y. Osaka, "Preparation and Properties of $In_xGa_{1-x}As$ Microcrystallites Embedded in SiO_2 Glass Films," *Jap. J. Appl. Phys.* 30: 136-142 (1991).

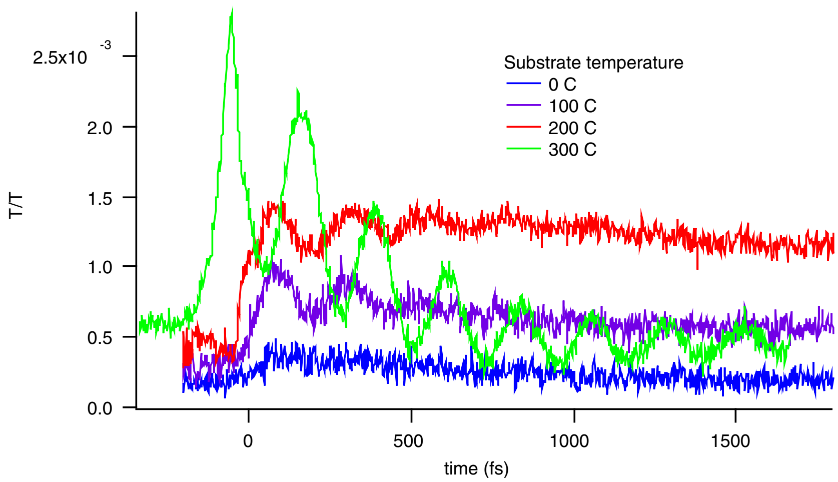

Figure 7: Coherent phonon generation in GaSb+SiO_2 films. Films were grown at 100 W rf power for 30 min and the substrate temperature was varied from 0°C to 300°C.

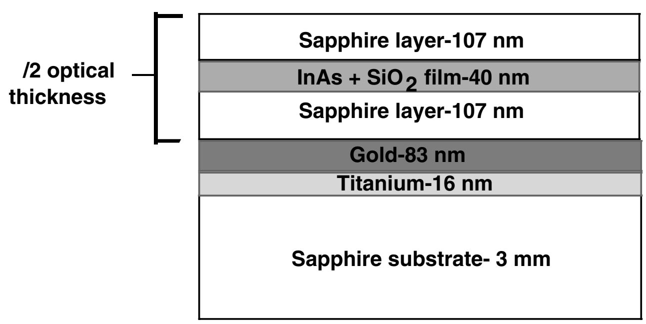

Figure 8: Design of a reflective saturable absorber device with sapphire spacer layers on a gold mirror.

The reflective saturable absorber devices consisted of a 40 nm thick InAs-doped silica film sandwiched between two 107 nm thick sapphire spacer layers and deposited upon a gold mirror (Figure 8), all grown in a non-magnetron rf sputtering system. The structures were fabricated and found to have good optical quality. Experiments aimed at characterizing and testing these devices are underway. In conclusion, non-epitaxially grown semiconductor-doped silica saturable absorber films have been shown to be an attractive alternative to epitaxially grown saturable absorbers. Improvements in this technology should enable its use in many applications requiring a compact, inexpensive source of ultrashort laser pulses.

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