Advances in Ultrafast Solid State Lasers

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ABSTRACT

An overview of the basic principles, major theoretical aspects and technological issues of self-mode-locked solid state lasers is presented, with particular emphasis on recent advances in femtosecond Ti:sapphire lasers.

Keywords: femtosecond phenomena, Ti:sapphire laser, ultrashort pulses, dispersion, mode-locking

1 INTRODUCTION

The last few years have brought about significant advances in ultrashort-pulse laser physics. The development of novel all-optical modulation techniques along with the appearance of ultrabroad-band solid state gain media opened up a new era in femtosecond pulse technology. More than two decades after the first demonstration of picosecond pulse generation in a solid state laser using a saturable absorber,[1] we have recently witnessed the emergence and evolution of a new generation of ultrashort-pulse lasers based exclusively on solid state components. This paper provides a brief synopsis of the basic operation principles and the major performance limitations of femtosecond solid state lasers (for a deeper insight and extensive reference lists the reader is referred to[2–4]), and summarizes recent technological advances allowing the generation of nearly bandwidth-limited sub-10-fs optical pulses directly from laser oscillators.

Although the first picosecond optical pulses were generated in a solid state (Nd:glass) laser in the Mid-60es,[1] progress in ultrashort-pulse solid-state laser technology came to a standstill shortly after the first pioneering experiments. It was the picosecond relaxation time of the saturable absorbers used for passive mode locking that prevented researchers from pushing the pulse duration below the picosecond limit in Nd:glass (and some other) solid state lasers. Hence interest shifted to organic dye lasers,[5] which, as a result of their nanosecond upper-state lifetimes, were capable of actively participating in short pulse formation,[6] allowing intracavity pulse shortening down to the femtosecond regime by using "slow" (picosecond)-relaxation-time absorbers.[7] Inspite of the relatively simple concept of slow-saturable-absorber mode locking, however, Operation of femtosecond dye

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lasers continued to be a highly sophisticated art up to the present day, mainly because of the large number of inaccessible system parameters. and the short-lived dye components. One of the demands of highest priority for a variety of (potential) application fields: Reproducibility of femtosecond laser performance could not be met until the recent development of femtosecond solid state systems.

2 THEORETICAL CONSIDERATIONS

The emergence of a femtosecond solid state laser technology has been possible by utilizing the intensity-induced change in the refractive index of a transparent insulating material (e.g. the laser host crystal), which essentially instantaneously follows the variation of the optical field intensity. This ultrafast Kerr-effect can be transformed into an almost instantaneous-response saturable absorber effect by introducing appropriate linear optical components into the cavity. The two most successful embodiments of this general concept have been *additive-pulse mode locking*,[8] and *self (or Kerr-lens) mode locking*.[9] The effect of these passive Kerr modulators on the envelope of the electric field $v(t)$ (e.g. on the optical axis) can to a first approximation be written as,[2,3]

$$\Delta v(t) = (\kappa/2 + i\phi)|v(t)|^2, \tag{1}$$

where κ and ϕ measure the strengths of self-amplitude and self-phase modulation (SAM & SPM), respectively. Other than SAM and SPM, intracavity group delay dispersion (GDD) is a major pulse shaping effect, which is characterized by the parameter

$$D = \left(\frac{\partial T_r(\omega)}{\partial \omega}\right)_{\omega=\omega_0}, \tag{2}$$

i.e. by the first derivative of the cavity round-trip time T_r with respect to the optical frequency. at the center of the laser oscillation spectrum. In order to avoid a strong pulse broadening due to the interaction of a pulse carrying a positive chirp (as a consequence of $\phi > 0$) with normal dispersion ($D > 0$) the intracavity GDD must be negative.

3 SELF STARTING CONDITIONS FOR PASSIVE MODE-LOCKING

Efficient SAM with $\kappa > 0$ is an indispensable requirement for the formation of a single mode-locked pulse out of the free running laser oscillation. The SAM coefficient κ must exceed a threshold value in order that the saturable absorber effect can overcome dynamic gain saturation.[10] This necessary condition is easily satisfied in most solid state lasers, except perhaps a few color center gain media having high emission cross sections. A sufficient condtion is, that the product of κ and the cw circulating intracavity power P_{av} need to exceed a critical value in order that the most intense initial fluctuation can evolve into a mode-locked pulse before it is disrupted due to the finite mode coherence time τ_{coh} in the free-running laser.[11] This can be written as:

$$\kappa P_{av} > \frac{1}{\ln(m_i)} \frac{T_r}{\tau_{coh}} \tag{3}$$

where T_r stands for the cavity round trip time and m_i for the number of the initially oscillating longitudinal modes. The effective mode coherence time is defined by the inverse FWHM of the first beat note of the free running laser. Meeting the second criterion is often difficult and thus additional mechanisms (e.g. active modulation, synch-pumping, real saturable absorber[12]) may be required for starting the mode-locking process. Unfortunately the use of these starting mechanisms adds complexity and often impairs the steady-state mode locking performance. Hence, in laboratory systems one often resorts to a most simple method: with optimized SAM uninterrupted self

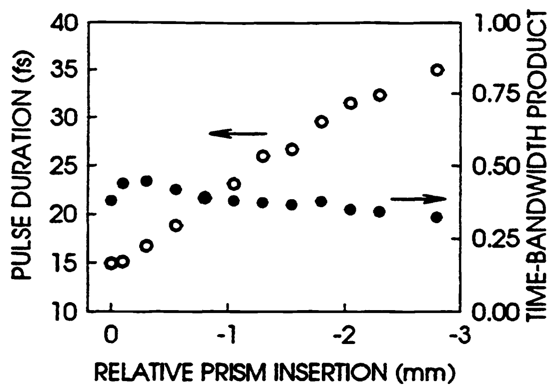

Fig. 1: Pulse duration and time-bandwidth product as a function of glass insertion in a fused-silica-prism-controlled Ti:S laser for a prism separation of 74cm and an intracavity pulse energy of 40 nJ. 1mm glass insertion corresponds to a GDD of $\approx$-95fs^2. At the zero refernce point the net negative GDD is -110$\pm$40fs^2.

mode locking operation over many hours can be achieved after starting the passive pulse formation by tapping one of the resonator mirrors.

4 STEADY STATE PASSIVE MODE LOCKING

Both additive-pulse and Kerr-lens modulators exhibit SPM coefficients that are much larger than the corresponding SAM parameters. $\phi >> \kappa$ implies that steady-state pulse formation is dominated by a soliton-like interplay between SPM and negative GDD. Hence, assuming i) a linear variation of $T_r(\omega)$ as a function of ω, i.e. $D(\omega) = D(\omega_0)$, over the mode-locked spectrum, ii) evenly distributed SPM and GDD in the cavity, and iii) a sufficiently broadband gain medium, the pulse duration τ is expected to approximately obey the well-know soliton formula

$$\tau \approx \tau_s = \frac{3.53|D|}{\phi W}, \tag{4}$$

where W stands for the intracavity pulse energy. The soliton-like behavior of the steady-state pulse has been verified in a self-mode-locked (quartz-prism-controlled) Ti:sapphire laser down to the 10 fs regime, as is shown in Fig. 1. Although κ does not significantly affect τ, optimized SAM is essential for stabilizing the pulse against noise and perturbations as well as preventing the emergence of a narrow-band cw background, which tends to co-exist with the mode-locked pulse in soliton-like (solitary) systems under non-optimized conditions.[4,13]

5 PERFORMANCE LIMITATIONS

In femtosecond solid state lasers the pulse duration can be reduced by decreasing the magnitude of negative intracavity GDD until one of the assumptions or approximations i)-iii) leading to Eq. (3) fails. The most severe limitation in practical broadband solid-state lasers (e.g. Ti:sapphire, Cr:LISAF,[15] Cr:forsterite[16]having bandwidths of the order of 100 THz) originates from the increasing deviation of $T_r(\omega)$ from a linear function as the oscillation spectrum broadens. The lowest-order contribution to this deviation causes a linear variation of $D(\omega)$ with frequency and is referred to as third-order dispersion (TOD). As the pulse duration is decreased and/or the pulse energy is increased the separated action of GDD and SPM increasingly modulates the pulse parameters

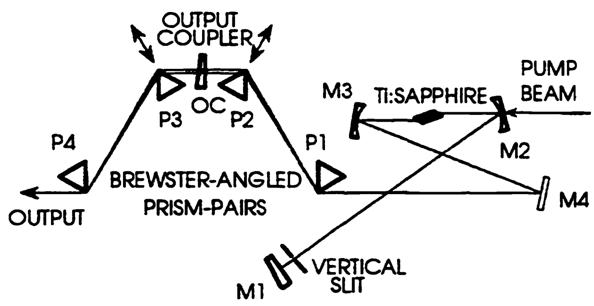

Fig. 2: Schematic of a prism-controlled self-mode-locked (hard aperture) Ti:S laser. For more details see Ref. 4.

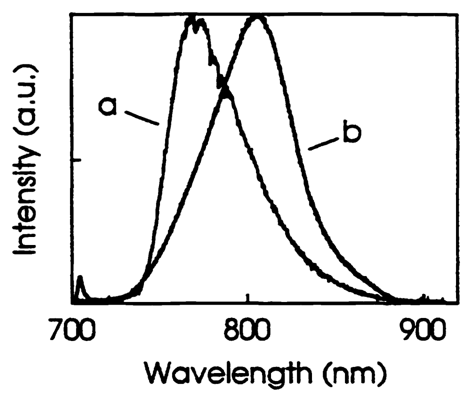

Fig. 3: Spectra obtained with (a) a prism-pair-compensated, and (b) with a MDC Ti:S laser.

(duration, bandwidth) as the pulse circulates in the cavity.[2] This modulation can be regarded as a periodic perturbation to the ideal soliton-like pulse and manifests itself in an additional term $\Delta\tau = \alpha(z)\phi W$, where α depends on the position in the cavity.[2] Other potential limitations to pulse shortening are the gain and resonator bandwidths, which, however, have turned out to be insignificant in practical systems so far. For a more detailed and quantitative analysis of the major effects limiting the performance of practical systems the reader is referred to Ref. 4. The considerations presented above generally apply to both additive-pulse and self mode-locking. The former is considered the most powerful technique for ultrashort pulse generation in *fiber* lasers, whereas self mode-locking is far superior to any other techniques if femtosecond pulses from *bulk* lasers are to be generated. In what follows we concentrate on self-mode-locked systems and refer to two recent reviews of APM fiber lasers.[17,18]

5.1 Prism-dispersion-controlled systems

Until recently broadband negative GDD has been introduced in short-pulse laser oscillators almost exclusively by a pair of Brewster-angled prisms. The layout of e.g. a prism-controlled self-mode-locked Ti:sapphire laser is shown in Fig. 2. To obtain the shortest output pulse duration, the circulating pulse is coupled out of the cavity after traversing the dispersive delay line,[2,19,20] and the extracavity prism pair allows GDD control outside the cavity. Since the prism pair introduces also high-order dispersion, a careful selection of the prism material is

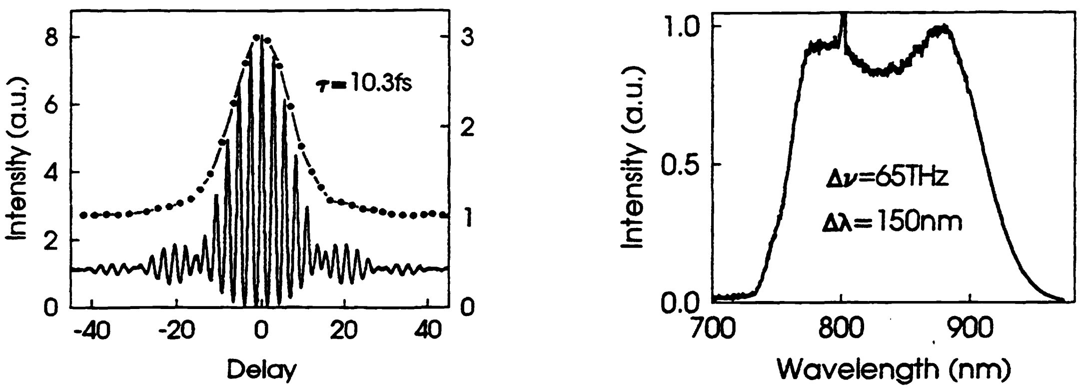

Fig. 4: Measured interferometric and evaluated intensity autocorrelation (left side). Note the disappearance of the structure in the wings of the intensity autocorrelation. Mode-locked spectrum of the fused silica prism pair controlled Ti:S laser around 850nm

needed if the pulse duration is to be minimized.[2] For Ti:sapphire the optimum choice turned out to be fused silica, which, by minimizing TOD in the cavity allowed the generation of nearly bandwidth-limited pulses of 11-12fs in duration around 0.8 μm.[22,23] The spectrum of a 12.3fs pulse[22] (trace (a) in Fig. 3) clearly exhibit a strong asymmetry as a result of residual negative TOD in the cavity, which sets a limit to further pulse shortening. Recent investigations[4,21] predicted a vanishing TOD in the Ti:sapphire/fused silica laser around 0.85 μm. As a matter of fact, tuning the laser to this wavelength extremely broad and yet symmetric mode-locked spectra (Fig. 4) can be generated. A careful evaluation[4] of the spectrum and the corresponding fringe-resolved autocorrelation (Fig. 4) yields, however, only a minor improvement in pulse duration because of the strong deviation of the evaluated pulse shape from a $sech^2$ profile. In this context it is important to notice that the intensity autocorrelation (upper trace in Fig. 4) shows little sensitivity to this deviation and hence inappropriate for a reliable and accurate determination of pulse parameters.
Nevertheless, at nearly zero GDD and TOD pulses as short as 8.7fs were generated.[20] In this operating regime the uncompensated fourth order dispersion was identified as the limiting factor for a further pulse shortening.[25]

5.2 Mirror-dispersion-controlled systems

Recently a novel technique has been proposed and demonstrated for intracavity dispersion control. Broadband, high-reflectivity multilayer dielectric mirrors have been developed with their multilayer period modulated during the evaporation process. This modulation not only broadens the high-reflectivity bandwidth but, more importantly, offers the possibility of engineering the dispersion properties of the mirrors.[26] As a first embodiment of this general concept, chirped dielectric mirrors exhibiting a nearly constant, i.e. high-order-dispersion-free, GDD over the wavelength range 720-890 nm ($\approx$ 80 THz) have been fabricated,[26] where the wavelength range can be easily shifted to match the emmision spectrum of other lasers by simply rescaling the layer thicknesses. Using these mirrors, compact, mirror-dispersion-controlled (MDC) self-mode-locked oscillators can be constructed, which are exemplified by the MDC Ti:sapphire laser shown in Fig. 5.[27] The significantly reduced TOD in this system as compared to its prism-controlled counterpart is clearly demonstrated by a symmetric $sech^2$-shaped spectrum in the 10fs domain (trace (b) in Fig. 3).

Corresponding to Eq. 4. the pulse duration is proportional to the GDD. In a MDC laser the net GDD can be only changed in discrete steps. But mirrors from different coating runs exhibit a slightly different GDD. A combination of mirors allow a fine tuning of the GDD.As revealed by Fig. 6, the optimized MDC Ti:sapphire laser is capable of producing highly stable, nearly bandwidth-limited 8 fs pulses.[28] This laser was pumped with by the blue green lines of a small frame argon laser. At 3 W absorbed pump power the cw and mode-locked output power with a 3.3% output coupler is 150–200mw and 60–100mW. With the resonator optimized for Kerr-lens-induced

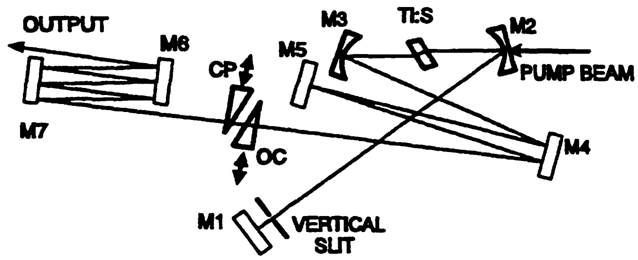

Fig. 5: Schematic of the MDC Ti:sapphire laser using chirped dispersive mirrors for intracavity (M4-M5) and extracavity (M6-M7) GDD control. M2,M3 are single stack quarterwave mirrors and OC the output coupler. With M6, M7 and a wedged compensating plate (CP) the extracavity GDD can be adjusted

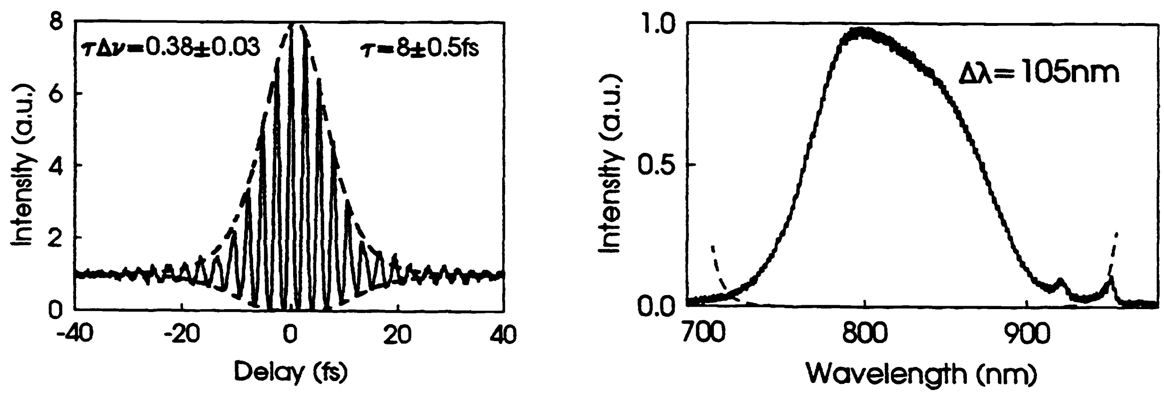

Fig. 6: Single scan, fringe resolved autocorrelation trace of the output of the MDC Ti:S laser. The dashed line represents the calculated envelope of an 8.2fs $sech^2$ -shaped pulse. Spectrum of the mode-locked laser (full line) and transmittivity of the dichroic mirrors (dashed line). The ends of the dashed lines correspond to a transmittivity of 10%.

amplitude modulation puls formation can be started by tapping one of the cavity mirrors and the laser stays mode-locked for many hours.

Special care has to be taken in steering and characterizing the broadband optical pulses exiting the mode-locked MDC laser. The spatial chirp and associated pulse front tilt induced by the wedged output coupler is eliminated by a second wedged glass plate identical to the output coupler substrate. This same glass plate (of varying thickness) also serves, in combination with chirped dispersive mirrors, for continuous tuning of the extracavity dispersion in the vicinity of a net zero GDD. The widely-used quarterwave dielectric beam splitter for p-polarized light is inadequate for distortion-free splitting of optical pulses with spectra extending over 200 nm. Hence, we rotate the originally horizontal polarization of the laser output by 90 degree with a pair of turning mirrors and use a broadband quarterwave beam splitter for s-polarization in the autocorrelator. This beam splitter exhibits less than 5% variation in the reflectivity and less than 0.5 fs variation in the reflected and transmitted group delay over the wavelength range of 650-950 nm. The dispersion of the 0.5-mm-thick fused silica beam splitter substrate is balanced by placing an identical AR-coated plate in the opposite arm of the autocorrelator. With the exception of the chirped mirrors and the beam splitter, the extracavity beam-steering and focusing optics comprise exclusively Au-coated unprotected mirrors. The pulses are focused by a mirror of 25 mm focal length onto a 25-μm-thick BBO frequency doubling crystal, in which the second-harmonic correlation signal is generated. Care has been taken to minimize the folding angle at the focusing mirror in order to keep astigmatism at minimum.

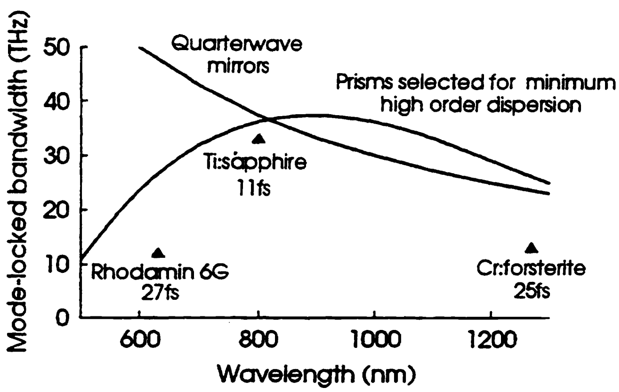

FIG. 7. Current state of the art bandwidth-limited pulse generation from laser oscillators

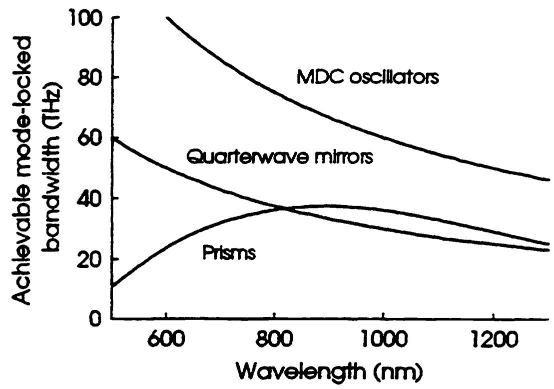

FIG. 8. Potential performance of mirror-dispersion-controlled and conventional femtosecond lasers

The fringe-resolved autocorrelation (FRAC) trace and the spectrum of the mode-locked laser output are shown in Fig. 6. A sech^2-fit to the measured FRAC-trace yields a pulse duration of $\tau = 8 \pm 0.5$ fs (full width of half maximum, FWHM) and a time-bandwidth product of $\tau\Delta\nu = 0.38 \pm 0.03$. Although the Fourier transform of the measured spectrum would give a pulse width of 8.6 fs in the absence of spectral phase modulation, this value is likely to be modified by the spectral response of the monochromator, which is not precisely known. Even with a constant spectral FWHM, the calculated minimum pulse width is utterly sensitive to slight changes in the wings of the spectrum. Regardless of the small uncertainty in the pulse width, the high visibility of the fringes in the wings of the autocorrelation trace and the absence of a substructure in the enevelope provide clear evidence for the high quality and the nearly transform-limited nature of the generated pulses.
A comparison of the mode-locked spectrum with the transmittivity of the quarterwave dichroic curved mirors (dashed line of Fig. 6) indicates that the major limitation preventing further pule shortening in the current system is the finite bandwidth the two curved mirrors. Because dispersion engineered mirrors can not be currently produced with a high transmittivity at the pump wavelength. A replacement of the dichroic focussing mirrors with broadband chirped mirrors calls for a new cavity configuration in which the output coupler serve as the input coupler for the input beam.
The maximum achievable mode-locked bandwith for conventional prism-pair dispersion compensated femtosecond oscillators is shown in Fig. 7. Whereas in the wavelength range below 850nm the high order dispersion of the prism pair is the limiting factor for a further pulse shortening, restricts over 850nm the high reflectivity bandwidth of standard dielectric mirrors the bandwidth. One way to overcome these limitations is the use of chirped mirrors. Beside the fact, that chirped mirrors are almost free from high order dispersion, they have also an extended bandwidth. The potential performance limitations of mirror dispersion controllled lasers in comparison to the conventional femtosecond lasers are shown in Fig. 8.

6 RELIABILITY AND REPRODUCIBILITY

In strong contrast to dye lasers, ultrashort pulse formation in solid-state lasers using Kerr-modulators can be described and controlled in terms of a few uniquely defined and experimentally easily accessible parameters (κ, ϕ, D, and W). As a result, the reliability of femtosecond solid-state lasers and the reproducibility of their performance is far superior to those of any previous femtosecond source. Whilst prism-controlled systems offer the advantage of continuously tunable intracavity dispersion, mirror-dispersion-controlled oscillators exhibit a cavity dispersion that is completely independent of resonator alignment, resulting in an unprecedented long-term stability and day-to-day reproducibility of femtosecond pulse parameters.

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8 REFERENCES

[1] A. J. DeMaria, D. A. Stetser, and A. Heynau, "Self mode locking of lasers with saturable absorbers," Appl. Phys. Lett. **8**, 174 (1966)

[2] F. Krausz, M. E. Fermann, T. Brabec, P. F. Curley, M. Hofer, M. H. Ober, Ch. Spielmann, E. Wintner, A. J. Schmidt, "Femtosecond solid state lasers," IEEE J. Quantum Electron. **QE-28**, 2097 (1992)

[3] E. P. Ippen, "Principles of passive mode-locking," Appl. Phys. B **B58**, 159 (1994)

[4] Ch. Spielmann, P. F. Curley, T. Brabec, F. Krausz, "Ultrabroad-band femtosecond lasers," IEEE J. Quantum Electron. **QE-30**, 1100 (1994)

[5] E. P. Ippen, C. V. Shank, and A. Dienes, "Passive mode locking of the cw dye laser," Appl. Phys. Lett. **21**, 348 (1972)

[6] G. H. C. New, "Mode locking of a quasi continuous laser," Opt. Comm. **6**, 188 (1972)

[7] R. L. Fork, B. I. Greene, and C. V. Shank, "Generatio of pulses shorter than 0.1ps by colliding pulse modelocking," Appl. Phys. Lett. **38**, 671 (1981)

[8] J. Mark, L. Y. Liu, K. L. Hall, H. A. Haus, and E. P. Ippen, "Femtosecond pulse generation in a laser with a nonlinear external cavity," Opt. Lett. **14**, 48 (1989)

[9] D. E. Spence, P. N. Kean, and W. Sibbett, "60-fsec pulse genaeration from aself mode locked Ti:sapphire laser," Opt. Lett. **16**, 42 (1991)

[10] E. P. Ippen, L. Y. Liu, and H. A. Haus, "Self starting conditions for additive-pulse mode-locked lasers," Opt. Lett.**15**, 183 (1990)

[11] F. Krausz, T. Brabec, and Ch. Spielmann, "Self starting passive mode locking,"Opt. Lett. **16**, 235 (1991)

[12] U. Keller, W. H. Knox, and G. W. t'Hooft, "Ultrafast solid state mode-locked lasers using resonant nonlinearities," IEEE J. Quantum Electron. **QE-28**, 2123 (1992)

[13] T. Brabec, S. M. J. Kelly, F. Krausz, "Passive mode locking in solid-state lasers," in *Compact Sources of Ultrashort Pulses*, Ed. I. N. Duling, Cambridge Press, 1994

[14] Y. F. Chou, J. Wang, H. H. Liu, and N. P. Kuo, "Measurement of the self-starting threshold of Kerr-lens mode-locking lasers," Opt. Lett. **19**, 566 (1994)

[15] N. H. Rizvi, P. M. W. French, and J. R. Taylor, "Generation of 33-fs pulses from a passively mode-locked Cr:LisaF laser," Opt. Lett. **17**, 1605 (1992)

[16] Y. Yanovsky, Y. Pang, F. Wise, and B. I. Minkov, "Generation of 25fs pulses from a self-mode-locked Cr:fosterite laser with optimized group-delay-dispersion," Opt. Lett. **18**, 1541 (1993)

[17] H. A. Haus, E. P. Ippen, and K. Tamura, IEEE J. Quantum Electron. **QE-30**, 200 (1994)

[18] M. E. Fermann, "Ultrashort pulse sources based on single-mode rare-earth-doped fibers," Appl. Phys. B **B58**, 197 (1994)

[19] B. Proctor and F. Wise, "Quartz-prism sequence for reduction of cubic phase in a mode-locked Ti:Sapphire laser," Opt. Lett. **17**, 1295 (1992)

[20] J. Zhou, G. Taft, C. Shi, H. C. Kapteyn, and M. M. Murnane, "Intracavity pulse duration measurement in a sub 10 fs Ti:sapphire oscillator," ICUP, paper MA2-1

[21] B. E. Lemoff and C. P. J. Barty, "Cubic-phase-free dispersion compensation in solid-state ultrashort-pulse lasers," Opt. Lett. **18**, 57 (1993)

[22] P. F. Curley, Ch. Spielmann, T. Brabec, F. Krausz, E. Wintner, and A. J. Schmidt, "Operation of a femtosecond Ti:sapphire laser in the vicinity of zero group-delay dispersion," Opt. Lett. **18**, 54 (1993)

[23] M. T. Asaki, C. P. Huang, D. Garvey, J. Zhou, H. C. Kapteyn, and M. M. Murnane, "Generation of 11-fs pulses from a self mode-locked Ti:sapphire laser," Opt. Lett. **18**, 977 (1993)

[24] J. Zhou, G. Taft, C. P. Huang, M. M. Murnane, H. C. Kapteyn, and I. Christov, "Pulse evolution in a broad bandwidth Ti:sapphire laser," Opt. Lett. **19**, 1149 (1994)

[25] I. P. Christov, M. M. Murnane, H. C. Kapteyn, J. Zhou, and C. P. Huang, "Fourth order dispersion limited solitary pulses," Opt. Lett. **19**, 1465 (1994)

[26] R. Szipöcs, K. Ferencz, Ch. Spielmann, and F. Krausz, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," Opt. Lett. **19**, 202 (1994)

[27] A. Stingl, Ch. Spielmann, F. Krausz, and R. Szipöcs, "Generation of 11-fsec pulses from a Ti:sapphire laser without the use of prisms," Opt. Lett. **19**, 204 (1994)

[28] A. Stingl, M. Lenzner, Ch. Spielmann, F. Krausz, and R. Szipöcs, "Sub 10 femtosecond, mirror-dispersion-controlled Ti:sapphire laser," Opt. Lett. **20**, March 15 (1995)