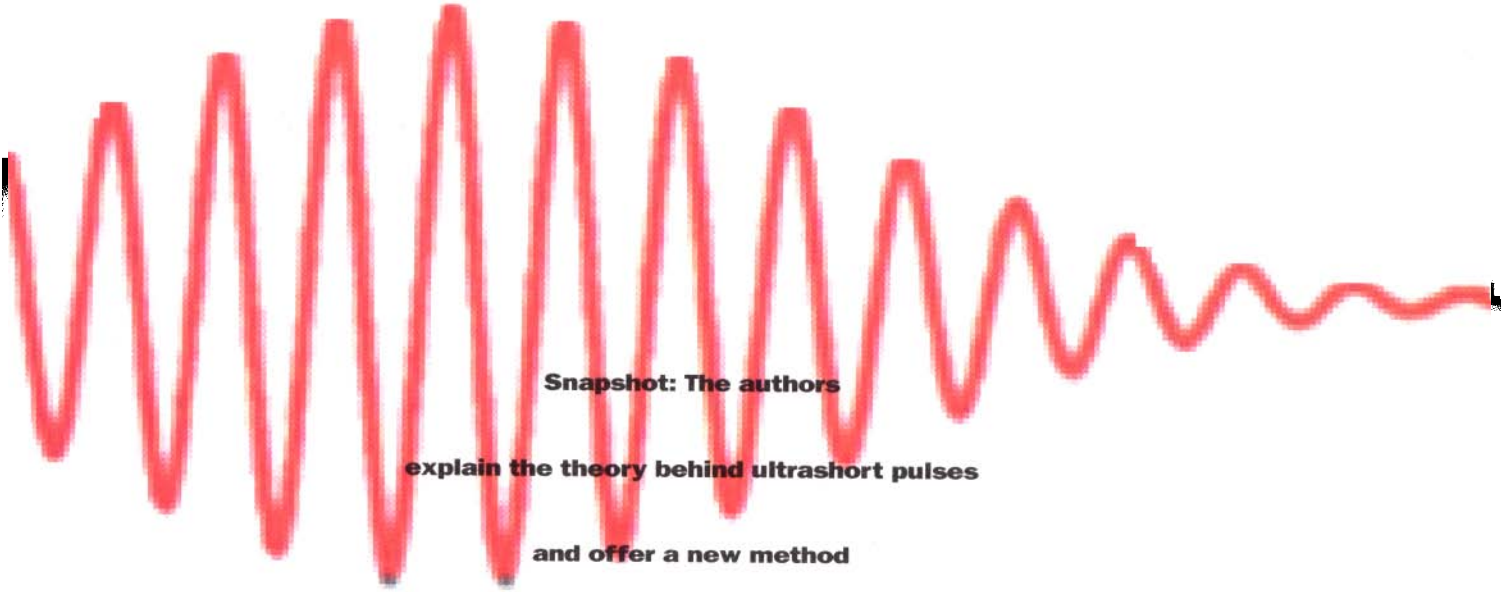
PUSHING THE LIMITS of Femtosecond Technology: Chirped Dielectric Mirrors

By Robert Szipöcs, Andreas Stingl, Christian Spielmann, and Ferenc Krausz

1047-6938/95/6/0016/05-$06.00



Snapshot: The authors explain the theory behind ultrashort pulses and offer a new method — chirped dielectric mirrors — for achieving sub-10 fsec pulses.

One of the major trends in laser physics today is the research and development of femtosecond laser sources. The motivation for generating short electromagnetic waveforms comes from many areas of science and technology. Ultrashort optical pulses are capable of taking "snapshots" of the state of matter and hence following the evolution of ultrafast processes at the microscopic level. Probing charge-carrier dynamics in semiconductors, the formation and breaking of chemical bonds, or time-resolved studies of photoisomerisation in biology are just a few examples of a number of intriguing applications. The generation of pulses shorter than previously possible would benefit many application fields and calls for a precise dispersion control over increasingly broad bandwidths in femtosecond laser oscillators as well as in subsequent optical systems. With the advent of chirped multilayer dielectric mirrors, feedback and phase dispersion control over unprecedented bandwidths have become feasible, thus opening the way for further advances in femtosecond technology.

Optical pulse propagation in dispersive media

Short electromagnetic waveforms (henceforth pulses) can be thought of as a superposition of long, quasi-monochromatic wavepackets of different carrier frequencies. For a particular spectral intensity distribution, minimum pulse duration is achieved when the centers of the wavepackets coincide in space. Under these conditions the pulse is referred to as Fourier-limited or transform-limited because the shortest pulse attainable with the spectral intensity distribution given has been realized.

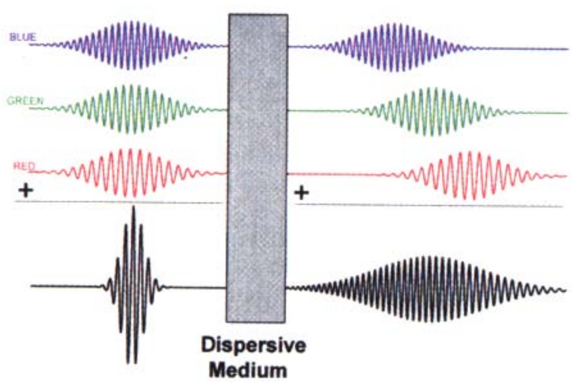


Figure 1. Optical pulse propagation through a dispersive medium: a frequency-dependent group delay leads to a pulse broadening and to a carrier frequency sweep.

If such a transform-limited pulse is sent through a dispersive medium, in which wavepackets of different carrier frequency propagate at different velocities, the pulse envelope will be broadened and carried by a time-varying instantaneous frequency at the output, as illustrated in Figure 1. Dispersion can be quantified by expanding the phase retardation $\phi(\omega)$ of a dispersive system about the center of the pulse spectrum ω_0 in the form

$$\phi(\omega) = \phi_0 + \phi'(\omega-\omega_0) + \tfrac{1}{2}\phi''(\omega-\omega_0)^2 + \tfrac{1}{6}\phi'''(\omega-\omega_0)^3 + \ldots \quad (1)$$

where $\phi_0 = \phi(\omega_0)$, and the derivatives ϕ', ϕ'', etc. are also evaluated at ω_0. The first derivative ϕ' is called group delay because it gives the time taken by the center of the pulse to reach the output of the wave-propagating medium. The higher-order terms in the expansion describe a frequency dependence of the group delay,

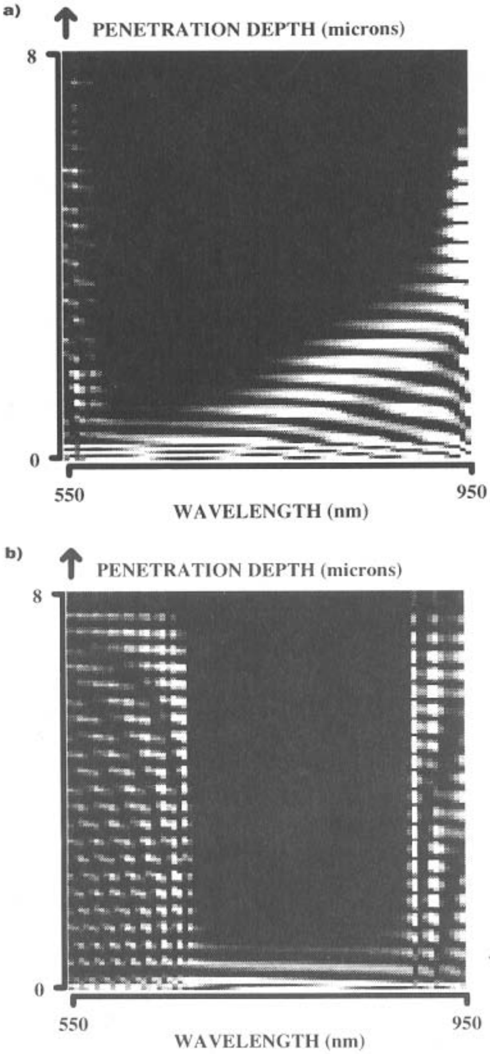


Figure 2. Electromagnetic energy-density distribution versus wavelength inside (a) a chirped multilayer dielectric mirror designed to provide a constant negative GDD and (b) a conventional dielectric mirror consisting of the same number of quarterwave layers.

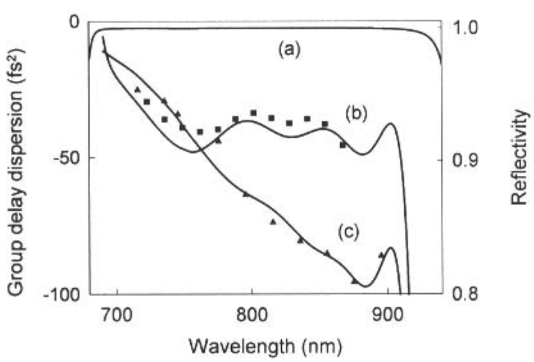


Figure 3. Curve (a): calculated reflectivity of chirped mirrors described in the text. The reflectivity is somewhat less (≈ 99.5%) in reality due to scattering losses not included in the calculations. Curves (b) and (c): calculated GDD of chirped mirrors designed for GDD and TOD compensation, respectively. The squares and triangles represent measured data points obtained with spectrally-resolved[15] and conventional[16] white-light interferometry, respectively.

and hence are responsible for dispersive effects. The lowest-order dispersion parameter ϕ'', which is termed group-delay dispersion (GDD), causes a broadening of the pulse without significant changes in the pulse shape and a carrier frequency varying linearly with time, which is often referred to as a linear chirp. The higher-order coefficients ϕ''' (third-order dispersion, TOD), ϕ'''' (fourth-order dispersion, FOD), etc. become only important for broad-bandwidth signals, and can give rise to a nonlinear chirp as well as to a distortion of the pulse. Critical values of the dispersion coefficients, above which dispersion causes a substantial change of the pulse, obey the simple scaling $\phi^{(n)} = \tau^n$, where τ is the pulse duration. For instance, a GDD of $\phi'' = \tau^2$ results in a pulse broadening by more than a factor of two. This scaling reveals a dramatic increase in the susceptibility to dispersion-induced broadening and distortion for decreasing pulse durations.

Dispersion in femtosecond laser oscillators

Ultrashort pulse generation from laser oscillators is ultimately limited by the finite oscillator bandwidth. However, both the gain bandwidth in organic dyes or vibronic solid-state gain media and the bandwidth of resonators formed by quarterwave multilayer dielectric mirrors support pulses much shorter than those produced previously. This is due to the fact that dispersive cavity components lead to a frequency-dependent cavity round-trip time (or group delay) $T_r(\omega)$, which restricts the "usable" bandwidth for ultrashort-pulse formation to a fraction of the full oscillator bandwidth. $T_r(\omega)$ can be expressed in terms of the dispersion parameters introduced above by deriving the Taylor series of the cavity-round-trip phase retardation $\phi_{cav}(\omega)$ as given by Eq. (1) with respect to ω.

The ideal $T_r(\omega)$ curve for optimum performance may be different in different femtosecond lasers due to differencies in the nonlinear optical processes responsible for pulse formation. In femtosecond dye lasers[1] oscillator-bandwidth-limited pulse generation requires a round-trip time nearly *constant* over the full oscillator bandwidth with a small amount of positive or negative cavity GDD (ϕ''_{cav}) depending on the operating conditions. The situation is quite different in the new generation of self-mode-locked femtosecond solid-state lasers.[2, 3] In these systems, the nonlinear index of the laser crystal (Kerr-effect) gives rise to a strong self-phase modulation of the circulating pulse. If a bandwidth-limited pulse interacts with the nonlinear index of a transparent optical medium, the spectrum of the pulse gets broadened. Frequency components lower than the carrier arise on the leading edge of the pulse whilst the higher-frequency components lag behind on the trailing edge. If such a *positively* chirped pulse experiences even a very small amount of positive cavity GDD ($\phi''_{cav}(\omega) > 0$), the leading edge completes every cavity round trip faster than the trailing edge of the pulse, resulting in a rapidly accumulating pulse broadening. As a consequence, it is imperative that the net cavity GDD be negative for femtosecond pulse generation in solid-state systems. A soliton-like interplay between self-phase modulation and *negative* GDD can shorten the circulating pulse to the limit set by the oscillator bandwidth, provided the GDD is constant over the full oscillator bandwidth. Despite the differences in the pulse forming mechanisms, what femtosecond lasers have in common, is that *higher-order cavity dispersion (TOD, FOD) must be negligible if oscillator-bandwidth-limited performance is to be achieved.*

The invention of prism pairs as a source of negative GDD[4] marked a milestone in the evolution of femtosecond laser technology. In dye lasers, intracavity prism pairs have provided a compensation of the positive material dispersion of intracavity components (*e.g.*, gain medium) and allowed continuous adjustment of the cavity GDD in a region of small positive and negative values. In a cavity formed by low-dispersion dielectric mirrors consisting of quarterwave layers[5], they yielded pulses as short as 27 fsec from a colliding-pulse mode-locked Rhodamine 6G (Rh6G) dye laser.[6] Prism-pair dispersive delay lines have also been key components of femtosecond solid-state lasers.[3] In these systems, the role of the prisms has been to overcompensate the positive dispersion of the laser crystal for soliton-like shaping.

The recent evolution of prism-controlled femtosecond solid-state lasers culminated in the development of a self-mode-locked Ti:sapphire laser using fused silica prisms,[7] which is capable of generating optical pulses of 10-15 fsec.[8-10] Both in the optimized Rh6G and Ti:sapphire laser, TOD of the prism pair[11, 12] has been identified as the major effect that prevents further pulse shortening as well as impairs the quality of the shortest pulses achieved. Approaching 10 fsec, an additional limitation tends to arise due to the finite bandwidth of the quarterwave mirrors. Hence, the evolution of ultrashort-pulse lasers has arrived at the frontiers of conventional femtosecond technology. Nevertheless, the gain linewidth of Ti:sapphire and related vibronic systems supports optical pulses as short as 3 fsec. These enormous bandwidths issue a great challenge to the community interested in the generation and application of ultrashort pulses. To rise to this challenge, novel techniques for providing feedback and dispersion control over ultrabroad bandwidths must be developed.

Chirped multilayer dielectric mirrors

The idea of chirped (aperiodic) refractive-index-modulated distributed-feedback (Bragg) structures dates back to the early 1970s, when Hill proposed the use of chirped Bragg reflectors for dispersion compensation in optical fiber systems.[13] The operation principle of these devices is fairly straightforward: different spectral components penetrate to different depths before reflected back from appropriate resonant sections of the chirped distributed-feedback structure. This introduces a frequency-dependent group delay, *i.e.* dispersion, which can be engineered. As an additional benefit, chirping the modulation period of a Bragg reflector also increases the bandwidth of its high-reflectivity (HR) range, just as a chirped pulse exhibit a broader frequency spectrum than its chirp-free counterpart.

These findings are directly applicable to an important special class of Bragg reflectors, namely multilayer dielectric mirrors. The widely-used standard quarterwave dielectric mirrors consisting of alternating quarterwave-layers of low- and high-index materials exhibit a low dispersion over a substantial part of their HR range.[5] Varying the layer thicknesses during the evaporation process provides a simple means of modulating the period of the multilayer structure.[14] These chirped multilayer dielectric mirrors (henceforth chirped mirrors) offer the benefits of a broader HR range (as compared to quarterwave mirrors) as well as an engineerable dispersion over the HR band. Consequently, the chirped mirror technology has the potential for overcoming the limitations researchers previously encountered in their attempts to produce shorter pulses from femtosecond oscillators.

To put these ideas into practice, chirped mirrors for use in Ti:sapphire oscillators and amplifiers have been designed and manufactured at the Research Institute for Solid State Physics Budapest (Hungary). The mirrors realized so far consist of 42 alternating quasi-quarterwave layers of SiO_2 (n_L=1.45) and TiO_2 (n_H=2.3) and have been produced by using standard electron-beam evaporation. For improving the performance of self-modelocked Ti:sapphire oscillators, chirped mirrors with a nearly constant negative GDD have been developed. The energy-density distribution as a function of wavelength inside the mirror is shown in Figure 2a. The most striking features are an increased HR bandwidth as compared to a quarterwave mirror shown in Figure 2b and a penetration depth that increases approximately linearly with wavelength between 700 and 800 nm. Curves (a) and (b) in Figure 3 show the calculated reflectivity and GDD of these mirrors, respectively. The HR range is currently limited by narrow resonances, which are not resolved in Figure 2a. Work is under way to eliminate these resonances. Nevertheless, even this "restricted" HR bandwidth is ≈30% broader than that of standard quarterwave mirrors. The negative GDD is approximately constant over

Fax Today! See page 62.

a bandwidth of ≈ 80 THz, opening the way for bandwidth-limited sub-10 fsec generation from Ti:sapphire lasers. Previous attempts to compensate dispersion with thin-film structures used resonant effects with much narrower bandwidths.[17, 18]

The broad constant-GDD bandwidth comes at the expense of a low value of the negative GDD. This relates to the fact that the overall group delay difference (= the product of bandwidth and GDD) is limited by the physical thickness of the mirrors, which must be traded off against low scattering losses.[14] One reflection off the broadband-GDD mirror (≈ -40 fsec^2) compensates the material GDD of 1 mm fused silica or 0.7 mm sapphire.[19] Hence, material dispersion must be minimized before these devices can be used for dispersion control.

Large amounts of material in the beam path are often unavoidable inside or outside the oscillator. In this case one has to resort to prism pairs limiting system performance by some residual TOD as discussed in the preceding section. Chirped mirrors can also be designed and fabricated to compensate for TOD, as shown in Figure 3. These mirrors exhibit a nominal GDD of ≈ -50 fsec^2 at 790 nm and a TOD of ≈ $+100$ fsec^3 and in combination with low-dispersion prisms can provide FOD-limited dispersion control in Ti:sapphire systems containing sizable amounts of optical material.

Chirped mirrors with similar or other dispersion characteristics can be produced for any wavelength range, where suitable (transparent) layer materials are available.[20] It is hoped that more sophisticated coating techniques will allow the fabrication of thicker stacks with tolerable scattering losses. These will provide higher dispersion parameters and/or broader bandwidths in the future.

Mirror-dispersion-controlled femtosecond systems

One of the most interesting application of the chirped mirrors is dispersion control in broadband femtosecond solid-state laser oscillators. To this end, the GDD-compensating mirrors characterized by the dispersion curve (b) in Figure 3 have been used to construct a prismless Ti:sapphire oscillator with a net negative GDD. Seven bounces on the dispersive mirrors (in one round trip) are necessary to overcompensate the positive GDD of a highly-doped 2-mm-thick Ti:sapphire crystal. Owing to the short gain length, a pump power of 3 to 4 W in the blue-green spectral region is sufficient (*e.g.*, from a small-frame argon or a frequency-doubled neodymium laser) for stable femtosecond pulse generation. With a 3%-transmitting output coupler, this mirror-dispersion-controlled (MDC) self-mode-locked Ti:sapphire oscillator delivers nearly bandwidth-limited 8 fsec optical pulses with a pulse energy of ≈ 1 nJ.[21] Figure 4 shows the fringe-resolved autocorrelation trace of the laser output. To our knowledge, these are the shortest pulses ever produced from a laser oscillator. They contain 3 cycles within their full width at half maximum just as the 6 fsec pulses obtained by extracavity pulse compression at 620 nm.[22]

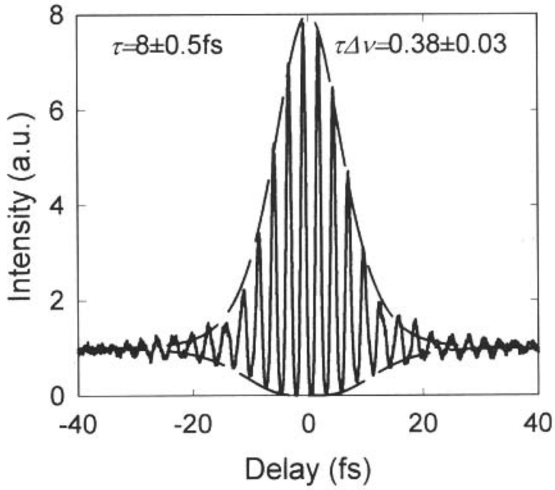


Figure 4. Interferometric second-order autocorrelation trace of the 8-fsec optical pulses generated by the mirror-dispersion-controlled Ti:sapphire oscillator described in Reference 21. The dashed-line around the pulse shows the calculated envelopes of an 8.2-fsec sech^2-shaped pulse.

The excellent stability of these sub-10-fsec pulses ≈1 % second harmonic fluctuations) offers an unprecedented sub-femtosecond resolution for ultrafast spectroscopy.[23] In strong contrast to prism-controlled Ti:sapphire systems producing pulses in this time domain,[12, 24] the 8 fsec pulses are generated at 800 nm, the center of the Ti:sapphire gain line, with a time-bandwidth product <0.4. These features make MDC Ti:sapphire oscillators ideal sources for both high-resolution ultrafast spectroscopy and seeding ultrashort-pulse Ti:sapphire amplifiers. The TOD-compensating chirped mirrors curve (c) in Figure 3 have recently been applied in combination with a pair of low-TOD prisms for recompressing the output of a kHz Ti:sapphire amplifier seeded with an MDC oscillator. This system currently produces 17-18 fsec pulses of an energy of 0.1mJ at a repetition rate of 1kHz.[25] These are the shortest optical pulses produced at multigigawatt power levels so far.

Conclusions

The availability of chirped dielectric mirrors permits the construction of more compact, stable, and user-friendly femtosecond oscillators and amplifiers than previously possible. In addition, they offer the potential for obtaining gain-bandwidth-limited pulses from Ti:sapphire and related broadband femtosecond solid-state oscillators. This means that the way is open toward the generation of optical pulses comprising only 1 cycle of light. We are entering exciting times in ultrafast science.

Acknowledgements

We wish to thank A. J. Schmidt and N. Kroo for their stimulating support and K. Ferencz for manufacturing the chirped mirrors. This research was sponsored by the Austrian and Hungarian *continued on page 59*

4-K—Ph.D. (1982) in Nonlinear Optics. Seeking a R&D/academic position in fiber optics, nonlinear optical devices, or femtosecond laser technology. Post-doctoral experience with above fields in an industrial R&D, university, and Govt. laboratory for more than 12 years (in Japan five years). Efficient in BASIC, FORTRAN, Apple/Macintosh, SUN/UNIX. More than 60 journal papers and conference presentations. Pioneer in characterizing thermo-optic coefficients of all optical materials in a new model. Fluent in Japanese.

4-L—Ph.D. (1990) in Engineering Physics. Seeking an academic or industry position related to fiber optic communications. Has been a project leader of nationally funded research projects since 1992. Has pursued research in large industry-related fiber-optic consortia. Proficient in fiber-optic system experiments, picosecond and femtosecond laser diodes, erbium-doped fiber amplifiers, and fiber gratings. Has been teaching undergraduate courses in optics and electronics. More than 30 publications.

4-M—Ph.D. (1980) in Optics. 20+ years academic and R&D experience in laser physics, nonlinear optics, harmonic generation, electro-optics, polarization spectroscopy and radiometry, fiber optics, medical laser applications. Design and development of solid-state lasers for differeent applications. Extensive experience in diode, excimer, and dye lasers, high vacuum system, passive Q-switch fabrication. Project management background. Multidisciplinary experience in physics research. Skilled in grants and proposals writing. Excellent teaching experience. Familiar with MathCad, C, interfacing and data acquisition. Nearly 40 publications. Willing to relocate. Not US citizen.

3-A—B.S. (1978) in Optics. With experience involving: FLIRs, fire control, guided munitions, night vision, lasers/LRFs, detectors, thin films, fiber optics, optical blacks, measurements, signatures, metrology, optical testing/alignment, interferometry, test equipment, experiments, breadboards/prototypes, BR/TDF, TIS, stray light/scatter, HEL beam control, radiometry/photometry, analysis, optical engineering, RDT&E EO systems, EO/IRCM/CCM, systems engineering, tight cost control, project management, client contact. Willing to relocate. U.S. citizen.

3-B—Ph.D. (1995), M.S. (1986) in Optics. Seeking a management, marketing, or R&D position in color science or optical industry. Especially interested in company having (or going to have) business (manufacturing or marketing) in China or Hong Kong. Previous management and marketing experience. Experience in photometry, colorimetry, and laser technology. Lived in China and in the U.S. and can comfortably build personal and business relationships in both locations. Permanent resident of U.S. Fluent in both Chinese and English.

3-C—Ph.D. (1995) in Applied Science, M.S. (1989) in Physics (Optics). Seeking industrial R&D position or national lab/academic post-doctoral position. Eight years industrial/national lab experience in optical characterization of materials, including nanocrystalline silicon for integrated optical devices, fullerene thin films for optical switching, wide bandgap nonlinear optical materials for harmonic generation, and thin magnetic films for RCS applications. Extensive experience (including development) with high power (YAG, YLF) and ultrafast laser systems (dye, Ti:sapphire) for materials characterization. Programming/modeling experience on both PCs and UNIX-based systems. Six journal publications and 11 conference presentations. U.S. citizen; DOE Q-clearance.

Pushing the Limits of Femtosecond Technology

Continued from page 20

Science Foundations, under grants P-9710, P-10409, and T-7376, respectively.

References

1. R. L. Fork *et al.*, "Generation of pulses shorter than 0.1 ps by colliding pulse modelocking," Appl. Phys. Lett. **38,** 671-673, (1981).
2. D. E. Spence *et al.*, "60-fsec pulse generation from a self-mode-locked Ti:sapphire laser," Opt. Lett. **16,** 41-43, (1991).
3. F. Krausz *et al.*, "Femtosecond solid-state lasers," IEEE Quantum Electron. **28,** 2097-2122, (1992).
4. R. L. Fork *et al.*, "Negative dispersion using pairs of prisms," Opt. Lett. **9,** 150-153, (1984).
5. S. De Silvestri *et al.*, "The role of cavity dispersion in cw mode-locked lasers," J. Quantum Electron. **20,** 533-539, (1984).
6. J. A. Valdmanis and R. L. Fork, "Design considerations for a femtosecond pulse laser balancing self phase modulation, group velocity dispersion, saturable absorption, and saturable gain," IEEE Quantum Electron. **22,** 112-118, (1986).
7. Ch. Spielmann *et al.*, "Generation of sub-20 fs mode-locked pulses from a Ti:sapphire laser," Electron. Lett. **28,** 1532-1533, (1992).
8. P. F. Curley *et al.*, "Operation of a femtosecond Ti:sapphire solitary laser in the vicinity of zero group-delay dispersion," Opt. Lett. **18,** 54-56, (1993).
9. B. Proctor and F. Wise, "Generation of 13-fs pulses from a mode-locked Ti:Al2O3 laser with reduced third-order dispersion," Appl. Phys. Lett. **62,** (1993) 470-473,.
10. M. T. Asaki *et al.*, "Generation of 11-fs pulses from a modelocked Ti:sapphire laser," Opt. Lett. **18,** 977-979 (1993).
11. B. E. Lemoff and C. P. J. Barty, "Cubic-phase-free dispersion compensation in solid-state ultrashort-pulse lasers," Opt. Lett. **18,** 57-59, (1993).
12. Ch. Spielmann *et al.*, "Ultrabroad-band femtosecond lasers," IEEE Quantum Electron. **30,** 1100-1114, (1994).
13. K. O. Hill, "Aperiodic distributed-parameter waveguides for integrated optics," Appl. Opt. **13,** 1853-1858, (1974).
14. R. Szipöcs *et al.*, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," Opt. Lett. **19,** 201-203, (1994).
15. A. Kovacs *et al.*, "Group-delay measurement on laser mirrors by spectrally resolved white-light interferometry," Opt. Lett. **20,** 788-789 (1995).
16. W. H. Knox *et al.*, "Interferometric measurements of femtosecond group delay in optical components," Opt. Lett. **13,** 574-576, (1988).
17. J. Heppner and J. Kuhl, "Intracavity chirp compensation in a CPM laser using thin film interferometers," Appl. Phys. Lett. **47,** 453-455, (1985).
18. J. M. Jacobson *et al.*, "Femtosecond pulse generation in a Ti:Al_2O_3 laser using second- and third-order intracavity dispersion," Opt. Lett. **17,** 1608-1610, (1992).
19. This should be contrasted with the orders of magnitudes higher GDD achievable with in-fiber Bragg reflectors. The bandwidth of these devices is, however, by orders of magnitudes smaller because of the much weaker index modulation. For details see e.g. K. O. Hill *et al.*, " Chirped in-fiber Bragg gratings for compensation of optical-fiber dispersion," Opt. Lett. **19,** 1314-1316, (1994).
20. Recently, chirped mirrors have been used for dispersion control in a femtosecond infrared OPO: J. Hebling *et al.*, "Chirped-mirror dispersion compensated optical parametric oscillator," accepted by Opt. Lett.
21. A. Stingl *et al.*, "Sub-10-femtosecond, mirror-dispersion-controlled Ti:sapphire laser," Opt. Lett. **20,** 602-604 (1995).
22. R. L. Fork *et al.*, "Compression of optical pulses to six femtoseconds by using cubic phase compensation," Opt. Lett. **12,** 483-485, (1987).
23. Ch. Spielmann *et al.*, "Tunneling of optical pulses through photonic band gaps," Phys. Rev. Lett. **73,** 2308-2311, (1994).
24. P. Zhou *et al.*, "Pulse evolution in a broad-bandwidth Ti:sapphire laser," Opt. Lett. **19,** 1149-1151, (1994).
25. Lenzner *et al.*, "Sub-20-femtosecond, kHz-repetition-rate Ti:sapphire amplifier," accepted by Opt. Lett.

R. Szipöcs is a postdoctoral researcher with the Research Institute for Solid State Physics Budapest, Hungary. A. Stingl and Ch. Spielmann are postdoctoral researchers, and F. Krausz is asstistant professor, in the Department of Quantum Electronics & Laser Technology, Technical University Vienna, Austria.