Sub-10-fs mirror-dispersion-controlled Ti:sapphire laser

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We demonstrate the generation of nearly bandwidth-limited 8-fs optical pulses near 0.8 μm from a self-mode-locked Ti:sapphire laser oscillator, using chirped dielectric mirrors for dispersion control. The mode-locking performance is described, and limitations are discussed.

The motivation for generating shorter, more intense, and higher-quality optical pulses comes from many fields in physics and, indirectly, from other areas of science and technology. Optical excitation (and probing) on a time scale of 10^{-14} s provides the only means of creating a coherent macroscopic polarization and investigating important related phenomena in semiconductors[1] as well as studying the dynamics of a number of chemical reactions or biological processes directly in the time domain. On the other hand, the availability of intense optical pulses approximately 10^{-14} s in duration should open the way to reversible nonlinear-optical experiments at intensity levels many orders of magnitude higher than previously feasible in the picosecond and subpicosecond time domains.[2,3]

In this Letter we report on what is to our knowledge the first ultrashort-pulse laser oscillator producing nearly bandwidth-limited optical pulses less than 10 fs in duration. The extremely broad bandwidth necessary to generate electromagnetic energy in such short intervals is provided by Ti-doped sapphire,[4] one of the most excellent broadband laser materials available these days. Exploitation of a substantial part of the enormous bandwidth of Ti:sapphire ($\approx$100 THz) has become possible with the discovery of self-mode locking (also termed Kerr-lens mode locking),[5] a unique method in that a passive amplitude modulation with virtually instantaneous response is introduced without the need for inserting any additional dispersive element in the cavity. Last but not least, femtosecond pulse formation in a broadband solid-state laser relies on a cavity round-trip time decreasing with the optical frequency near the gain line center [negative group-delay dispersion (GDD)] because of the strong self-phase modulation inherent in theses systems. In our self-mode-locked Ti:sapphire laser negative GDD is introduced by the same high-reflectivity dielectric mirrors that provide the feedback necessary for oscillation. These specifically designed chirped multilayer mirrors[6] can produce approximately constant negative GDD over a broader-bandwidth $\Delta\nu_{\mathrm{GDD}}$ than any other low-loss dispersive optical system demonstrated to date.

The finite value of $\Delta\nu_{\mathrm{GDD}}$ has been recognized to be a major effect limiting the performance of practical self-mode-locked oscillators. In systems employing prism pairs for controlling cavity dispersion,[7] third-order dispersion introduced by the prism pair itself was identified as the dominant limitation. Improved performance obtained by use of selected prism materials provided evidence for this finding.[8] The evolution of prism-controlled self-mode-locked Ti:sapphire lasers culminated with the development of a Ti:sapphire oscillator using fused-silica prisms,[9] which were shown to introduce the lowest cubic phase distortion near 0.8 μm among the commercially available optical materials. These systems can now produce pulses in the 10–15-fs region[10–12] but are still limited by the residual cavity third-order dispersion, as evidenced by their pronounced spectral asymmetry.

More recent investigations revealed that the overall cavity third-order dispersion vanishes at $\approx$0.85 μm in a Ti:sapphire laser using fused-silica prisms.[13,14] In fact, extremely broad double-peaked spectra centered at 0.85 μm could be obtained.[14,15] However, the increase in mode-locked bandwidth (beyond 150 nm) was not accompanied by a corresponding reduction in pulse duration because of fourth-order dispersion of the prisms[16] and possible coherent ringing owing to detuning.[17] A careful spectral analysis and fit to the interferometric autocorrelation yielded pulse durations of $\approx$10 fs,[14] whereas 8.5–9-fs pulse durations were evaluated by the simple assumption of a sech^2 pulse shape.[14,15]

In summary, prism-controlled Ti:sapphire lasers are capable of generating high-quality nearly sech^2-shaped pulses with durations as low as $\approx$15 fs. Below 15 fs the spectrum tends to become increasingly asymmetric, and pulse durations near 10 fs can be achieved only at the expense of detuning and a poor pulse quality characterized by time–bandwidth products of $\approx$0.6. In contrast, the mirror-dispersion-controlled (MDC) Ti:sapphire laser presented here delivers 8-fs nearly sech^2-shaped pulses with approximately symmetric spectra and time–bandwidth products of $\approx$0.38.

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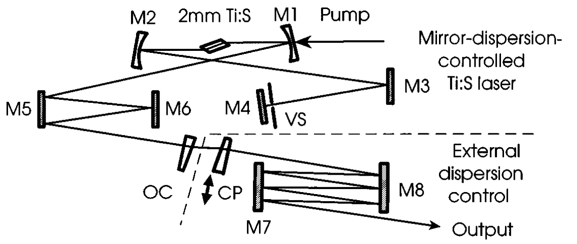

Fig. 1. Schematic of the MDC femtosecond Ti:sapphire (Ti:S) laser. The pump beam is focused with a 40-mm lens onto a 2-mm-thick highly doped Ti:S crystal (absorption coefficient, 5 cm^{-1} at 514 nm). M1, M2, single-stack quarter-wave dichroic mirrors highly transmitting at the pump wavelengths with radii of curvature of 5 cm; M3–M8, chirped dispersive mirrors; VS, vertical slit; OC, 3.3% broadband output coupler; CP, wedged glass plate.

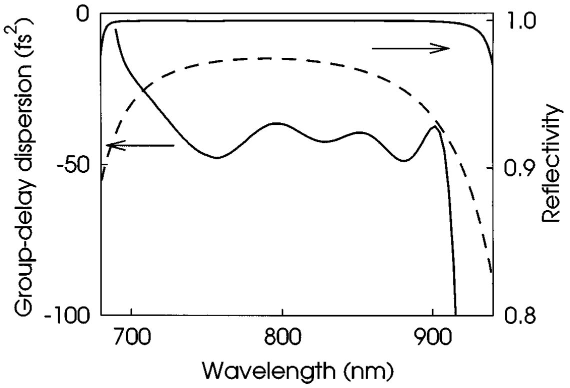

Fig. 2. Reflectivity and GDD of the chirped dielectric mirrors (solid curves) and the reflectivity of the output coupler (dashed curve).

A schematic of the MDC laser is shown in Fig. 1. To increase $\Delta\nu_{\mathrm{GDD}}$, we have redesigned the chirped multilayer mirrors used in a forerunner of the present system.[18] As revealed by Fig. 2, the new mirrors exhibit a $\Delta\nu_{\mathrm{GDD}}$ extending over ≈200 nm. The broader negative-GDD bandwidth, however, comes at the expense of a somewhat reduced value of average negative GDD. Because of the reduced mirror dispersion the number of reflections off the dispersive mirrors per round trip has been increased from 7 to 9 by the insertion of an additional folding mirror in the short collimated cavity arm. This has also resulted in a more balanced distribution of the negative GDD in the cavity, reducing the discreteness of solitonlike shaping and the related perturbations to the steady-state pulse.[14] However, this MDC cavity still has a drawback in that we are unable to control the net cavity GDD continuously. To improve this situation, we have adopted two different types of mirror with comparable bandwidths but with slightly different GDD that permit us to adjust the net cavity dispersion in small steps ($\Delta D \approx 10$ fs^2).

Because of the broad bandwidth of the chirped mirrors a conventional low-transmittivity quarter-wave output coupler would introduce a severe limitation to the oscillator bandwidth. Hence a specific broadband output coupler has been developed that not only exceeds the bandwidth of a quarter-wave output coupler having comparable transmittivity by some 30 nm (see the dashed curve in Fig. 2) but also introduces a broadband negative cubic dispersion for partial compensation of the positive cubic dispersion introduced by the laser crystal.

The laser is pumped by the blue-green lines (488 and 514 nm) of a small-frame Ar laser. At 3 W of pump power the cw and mode-locked output powers with the 3.3% output coupler are 150–200 and 60–100 mW, respectively. This mode-locked average output power corresponds to an output pulse energy of ≈1 nJ at a repetition rate of ≈80 MHz. Mode locking is accomplished in the usual manner[14]; i.e., the distance between the two curved mirrors is adjusted close to the upper boundary of the lower-stability region (corresponding to shorter mirror spacing), and a vertical slit is placed in the shorter collimated arm close to the end mirror. An optimum trade-off between stability and maximum self-amplitude modulation was achieved with a ratio of the lengths of collimated cavity arms of ≈2:1, yielding 0.45-mm-long stability regions 0.25 mm apart.[14] With the resonator optimized for Kerr-lens-induced amplitude modulation we can start pulse formation by tapping one of the cavity mirrors, and the laser stays mode locked without any notable degradation of performance for many hours.

We must take special care in steering and characterizing the broadband optical pulses leaving 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. The same glass plate (of varying thickness) also serves, in combination with chirped dispersive mirrors, to tune continuously the extracavity dispersion in the vicinity of a net zero GDD. The widely used quarter-wave 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° with a pair of turning mirrors and use a broadband quarter-wave 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. We balance the dispersion of the 0.5-mm-thick fused-silica beam-splitter substrate by placing an identical antireflection-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 are composed exclusively of Au-coated unprotected mirrors. The pulses are focused by a mirror of 25-mm focal length onto a 25-μm-thick beta-barium borate 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 to keep astigmatism at a minimum.

The fringe-resolved autocorrelation (FRAC) trace and the spectrum of the mode-locked laser output are shown in Fig. 3. A sech^2 fit to the measured

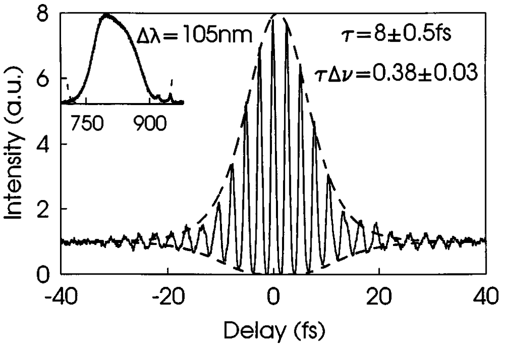

Fig. 3. Single-scan FRAC trace of the output of the MDC Ti:sapphire laser. The dashed curves represent the calculated envelopes of an 8.2-fs sech^2-shaped pulse. The collinear autocorrelator has been calibrated by use of a He–Ne laser. The fringe asymmetry is presumably caused by a slight bend of the thin beam splitter or the astigmatism introduced by the focusing mirror. Inset: spectrum of the mode-locked laser (solid curve) and transmittivity of the dichroic curved mirrors (dashed curve). The ends of the dashed curve correspond to a transmission of 10%.

FRAC trace yields a pulse duration of $\tau = 8 \pm 0.5$ fs (FWHM) and a time–bandwidth product of $\tau\Delta\nu = 0.38 \pm 0.03$.[19] The high visibility of the fringes in the wings of the FRAC trace and the absence of substructure in the envelopes provide clear evidence for the high quality and the nearly transform-limited nature of the generated pulses.

Although these optical pulses are still somewhat longer than the shortest pulses obtained by external pulse compression (6 fs),[20] they offer what we believe is an unprecedented time resolution for spectroscopy owing to their high stability and repetition rate. In a recent experiment subfemtosecond time resolution was demonstrated by use of 12-fs pulses generated by a forerunner of the present system.[21]

A comparison of the mode-locked spectrum with the transmittivity of the quasi-quarter-wave dichroic curved mirrors (dashed curve inset of Fig. 3) indicates that the major limitation preventing further pulse shortening in the current system is the finite bandwidth of the two standard quarter-wave cavity mirrors. Because dispersion-engineered mirrors cannot currently be produced with high transmittivity at the pump wavelengths, a replacement of the dichroic focusing mirrors with broadband chirped mirrors calls for a new pump configuration.

In conclusion, we have demonstrated what to our knowledge is the first laser oscillator that generates nearly bandwidth-limited high-quality optical pulses in the sub-10-fs regime. The mode-locked spectrum is centered close to the peak of the gain line in Ti:sapphire.[22] These features, along with the high stability and reproducibility of performance, make this system a unique tool both for high-resolution time-resolved spectroscopy and for seeding ultrashort-pulse Ti:sapphire amplifiers.

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