Femtosecond pulse generation from a synchronously pumped Ti:sapphire laser

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We report femtosecond mode locking of a Ti:sapphire laser synchronously pumped by a mode-locked, frequency-doubled Nd:YLF laser. Stable tunable 70-fs pulses are generated in a TEM_{00} output without the need for stabilization of the cavity length.

The study of ultrafast processes in physics and chemistry relies on the use of laser systems that produce stable ultrashort optical pulses. So far, the passively mode-locked dye laser has been the only widely used, reliable source of femtosecond light pulses.[1] Recently passive mode locking of visible and near-infrared solid-state lasers (Ti:sapphire[2-4] and Nd:glass[5-7]) led to stable femtosecond pulse generation, which opened a new era in the development of ultrafast technology. These new laser systems achieve superior performance and have reduced cost and complexity compared with traditional dye-laser technology. The lasers in Refs. 2-7 have been pumped by cw gas (argon, krypton) lasers. In this Letter we report femtosecond pulse generation in a Ti:sapphire laser synchronously pumped by a cw actively mode-locked, frequency-doubled Nd:YLF laser. Tunable bandwidth-limited pulses of 70-fs duration have been produced. To our knowledge this is the first solid-state laser system generating tunable femtosecond pulses in the visible and near-infrared regions of the spectrum.

Owing to its exceptionally broad fluorescence line, which extends over 400 nm, and to other favorable characteristics, Ti:sapphire is an attractive laser material for tunable ultrashort-pulse generation and amplification. Continuous-wave Ti:sapphire lasers have been actively mode locked by acousto-optic modulation[8-10] and synchronous pumping[11] to yield pulse durations of 1.3 ps (Ref. 9) and 200 ps,[11] respectively. Passive[2-4] mode-locking techniques have resulted in the production of femtosecond pulses as short as 60 fs.[3]

The experimental setup of the laser system described in this Letter is illustrated in Fig. 1. The lamp-pumped cw Nd:YLF laser (Quantronix 4216D) is mode locked by acousto-optic loss modulation and is capable of producing pulses shorter than 60 ps at a repetition rate of 76 MHz. The average output power is 20 W at the wavelength of 1053 nm. This radiation is frequency doubled by temperature-phase-matched second-harmonic generation (SHG) in a lithium triborate (LBO) crystal.[12] The crystal length and aperture measure 12 mm and 5 mm × 5 mm, respectively. The infrared beam is focused by lens L1 (focal length $f_1 = 10$ cm) into the crystal and recollimated with lens L2 ($f_2 = 15$ cm). Stable long-term operation with second-harmonic fluctuations of less than 4% (peak to peak) is achieved at a frequency-doubled output of 4 W, but the cavity length needs readjustment hour by hour to maintain this stability. Owing to the large angle acceptance of 90° phase matching in LBO (Ref. 13) and the high beam pointing stability of an YLF laser, the frequency-doubled laser output has an excellent beam quality, comparable with those of argon-ion lasers.

The Ti:sapphire laser is a five-mirror astigmatically compensated arrangement as shown in Fig. 1. The Ti:sapphire laser rod is cut at Brewster's angle with the *c* axis perpendicular to the propagation direction. The length of the crystal is 15 mm, and it absorbs 80% of the pump radiation at 527 nm. The pump beam is matched into the cavity through dichroic mirror M3. The focusing mirror M2 is highly reflecting at both pump and laser wavelengths. The resonator is approximately 2 m long, and its length can be adjusted by translating mirror M4 along the cavity axis. The SF10 Brewster prisms P1 and P2 allow a convenient control of the cavity dispersion. In our experiments, wavelength tuning has been accomplished by translating a variable-aperture slit perpendicular to the cavity axis between P2 and M4.

With 3% output coupling the threshold for cw laser oscillation is at ≈1 W of absorbed pump power, and the slope efficiency (output power versus absorbed pump power) is ≈10% when the laser is tuned to the maximum-gain wavelength. Synchronous mode locking is easily obtained by matching the cavity length to that of the pump laser. Using a fast photodiode and oscilloscope combination with a response time of 120 ps, one observes a stable pulse train well synchronized to the pump laser when the cavity length of the Ti:sapphire laser is matched to the repetition rate of the pump pulses to within a few micrometers. From this measurement we have concluded that the optical pulses have durations of less than 100 ps and a good energy stability. However, the second harmonic of the Ti:sapphire laser output appears to fluctuate strongly on a millisecond time scale at the same time. These observations along with autocorrelation measurements suggest

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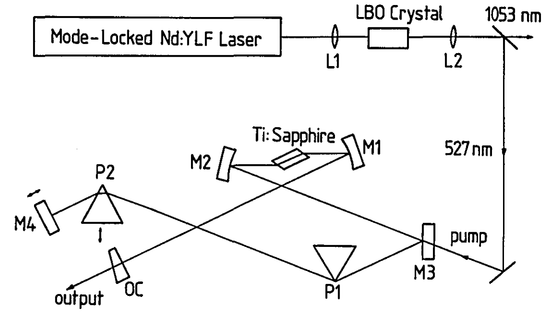

Fig. 1. Schematic diagram of the mode-locked laser system. M1, M2, concave spherical mirrors with $R = 12$ cm; M3, M4, plane mirrors; OC, plane output coupler with $T = 3\%$.

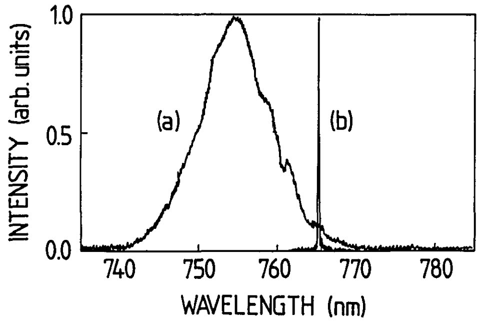

Fig. 2. Spectrum of the mode-locked Ti:sapphire laser for an absorbed pump power greater than 2.5 W [curve (a)] and less than 2.5 W [curve (b)].

that the duration (and presumably the shape) of the mode-locked pulse is subject to substantial fluctuations, with average pulse durations between 20 and 30 ps. The (averaged) spectrum of the synchronously mode-locked laser measured by a slowly scanning monochromator is 0.2 nm wide and centered at 765 nm, as shown in Fig. 2.

As the absorbed pump power is increased above 2.5 W, the noisy autocorrelation trace becomes clean and indicates a dramatic reduction of pulse duration. Once the second threshold is reached, synchronization between the sapphire laser and the pump laser ceases, and stable mode locking tolerates considerably more detuning, typically several tens of micrometers. This tolerance is dependent on pump power, with a maximum allowable detuning becoming greater with increased pump power. The desynchronization indicates that passive mechanisms take command over mode locking. However, synchronous pumping still plays an important role in the buildup and maintenance of stable mode-locked operation since detuning beyond the limits mentioned above results in a much greater sensitivity to external physical perturbations, and the femtosecond mode-locking process does not start spontaneously beyond these limits.

At an absorbed pump power of 3.2 W the average output power is 200 mW, and the optimum net round-trip group-delay dispersion, for which the shortest pulses are produced, has been found to be $D = \partial^2\phi/\partial\omega^2 = -4 \times 10^3$ fs^2. Under these conditions pulses as short as 65 fs have been generated, and the laser routinely produces pulses between 70 and 80 fs. A typical autocorrelation trace is shown in Fig. 3. For the determination of the pulse width a sech^2 pulse shape was assumed. The corresponding spectrum is shown in Fig. 2. It is blue shifted from the spectrum of the purely synchronously mode-locked oscillation with a center wavelength of 755 nm. A similar blue shift was observed in a passively mode-locked Nd-doped fiber laser recently.[14] The 10-nm-wide spectrum yields a time–bandwidth product $T_p\Delta\nu = 0.37$, which is close to the Fourier-transform limit of 0.32 for sech^2 pulses. The tuning range of the synchronously mode-locked laser with the present optics extends from 715 to 855 nm, but the laser ceases passively mode-locked operation outside the range between 740 and 800 nm because of the reduction of intracavity power.

It is important to notice that the laser requires a slight misalignment of the end mirrors to produce femtosecond pulses. However, the misalignment implies only a small (<10%) reduction of average output power and does not affect TEM$_{00}$ oscillation. This is in strong contrast to a self-mode-locked Ti:sapphire laser, for which a strong cavity misalignment, leading to a 40% reduction in output power and to higher-order transverse-mode oscillation, was necessary to achieve femtosecond pulse generation.[3] Even so, the laser was reported to be sensitive to external perturbations, and self-locking could be induced only by applying some external physical shock. In the synchronously pumped Ti:sapphire laser, however, femtosecond pulse generation is completely self-starting even at comparatively low (>2.5 W) pump powers, and interruption-free stable long-term operation is achieved with the laser mounted on a stainless-steel breadboard without any kind of environmental isolation. Stable femtosecond pulses with second-harmonic fluctuations of less than 15% are maintained over hours with slight readjustment of the cavity length of the pump laser from time to time.

The gain modulation in the synchronously pumped Ti:sapphire laser is less than 1% owing to the long upper-state lifetime of the gain medium (compared with that of dye lasers). As a consequence, the presence of efficient passive pulse-shaping mechanisms is absolutely necessary for femtosecond pulse forma-

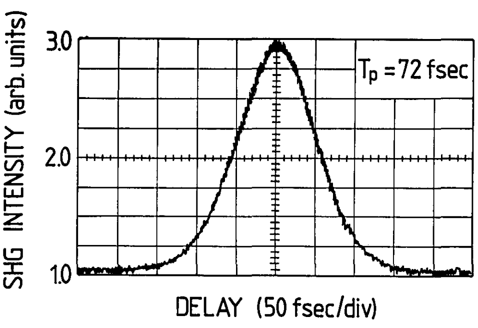

Fig. 3. Collinear autocorrelation trace of the mode-locked laser output corresponding to Fig. 2(a).

tion. Self-phase modulation by the Kerr nonlinearity of the gain medium in conjunction with negative dispersion is expected to play a dominant role in ultrashort-pulse evolution in the system presented here. If so, the pulse evolution in the cavity is governed by the nonlinear Schrödinger equation,[15]

$$\frac{\partial U}{\partial n} = i\Delta_D \frac{\partial^2 U}{\partial \tau^2} + i\Delta_{NL}|U|^2 U, \tag{1}$$

as long as the changes in pulse shape and phase in the discrete cavity elements are small and bandwidth limitation may be neglected. Here $U(n, \tau)$ is the normalized field amplitude, n is the number of round trips, and $\tau = t/T_0$ represents the normalized time variable, where T_0 is the initial pulse width (of the purely synchronously mode-locked laser). Further, $\Delta_D = |D|/2T_0^2$, and $\Delta_{\mathrm{NL}} = 2\gamma P_0 L_{\mathrm{NL}}$, where γ and L_{NL} represent the effective Kerr coefficient[15] and the length of the active medium, respectively, and P_0 stands for the initial peak power of the pulse. With the parameters of the synchronously mode-locked picosecond laser we obtain $\Delta_{\mathrm{NL}} \gg \Delta_D$, which implies a pulse evolution in which an initial pulse shortening is followed by a pulse-spreading phase, which leads to periodic fluctuations of the pulse shape reminiscent of high-order solitons.[15] This conclusion is supported by the observation of strong second-harmonic fluctuations produced by the picosecond output pulses of nearly constant energy.

It can be shown generally that pulse propagation through a system with distributed dispersion and distributed self-phase modulation is not capable of compressing a picosecond pulse into a steady-state femtosecond pulse. Therefore we have introduced some passive amplitude modulation to Eq. (1) by replacing Δ_{NL} with $\Delta_{\mathrm{NL}} - i\Delta_A$, where $\Delta_A = \kappa P_0$ (an ideal fast saturable absorber). If we do this the right-hand side of Eq. (1) has to be supplemented with a further term, $-\alpha(n)U$, that accounts for a change in the saturated round-trip gain during the pulse evolution. As the transition from the picosecond to the femtosecond mode-locked state occurs without significant change in pulse energy, $\alpha(n)$ has been determined from the requirement of constant pulse energy during the transient mode-locking process.[16] Our computer simulations have shown that, under these conditions, continuous pulse shortening down to a femtosecond steady state is possible if Δ_A reaches a certain threshold value. We surmise that $\kappa > 0$ originates from an intensity-dependent gain, which is the consequence of an intensity-dependent overlap between the pump and cavity beams owing to self-focusing in the gain medium. Cavity misalignment leads to a mismatch between pump and cavity mode and may increase κ under certain conditions. For a fixed value of κ, our calculations predict a threshold peak power P_0 for femtosecond pulse production, in full agreement with the experimental observation. Recently, a similar threshold was observed in a dispersion-controlled Ti:sapphire laser mode locked by a saturable absorber.[4]

The long-term stability of the presented system mainly depends on that of the pump laser. Stable hands-off operation may be obtained by employing feedback techniques that prevent the mode-locker drive frequency and cavity length from drifting apart in the pump laser.[17] Higher pump powers will extend the tuning range of the femtosecond pulses. As a further improvement, the lamp-pumped Nd:YLF laser can be replaced by a powerful diode-pumped laser in the near future.[18] Such a compact, long-lived, all-solid-state tunable femtosecond laser system has the potential of becoming the future workhorse for ultrafast spectroscopy.

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