Optics Communications 91 (1992) 77–81
North-Holland

OPTICS
COMMUNICATIONS

Generation of dual-wavelength, synchronized, tunable, high energy, femtosecond laser pulses with nearly perfect gaussian spatial profile

J.-K. Wang, Y. Siegal, C. Lü and E. Mazur
Gordon McKay Laboratory, Harvard University, Cambridge, MA 02138, USA

Received 13 January 1992; revised manuscript received 12 March 1992

We use self-phase modulation in a single-mode fiber to produce broadband femtosecond laser pulses. Subsequent amplification through two Bethune cells yields high-energy, tunable, pulses synchronized with the output of an amplified colliding-pulse-modelocked (CPM) laser. We routinely obtain tunable 200 μJ pulses of 42 fs (fwhm) duration with a nearly perfect gaussian spatial profile. Although self-phase modulation in a single-mode fiber is widely used in femtosecond laser systems, amplification of a fiber-generated supercontinuum in a Bethune cell amplifier is a new feature which maintains the high-quality spatial profile while providing high gain. This laser system is particularly well suited for high energy dual-wavelength pump = probe experiments and time-resolved four-wave mixing spectroscopy.

The rapid developments in nonlinear laser spectroscopy and ultrafast phenomena have created a need for laser systems satisfying stringent requirements. First of all, many four-wave mixing spectroscopic techniques require a synchronized source of two different frequencies ω_1 and ω_2. Furthermore, at least one of these two frequencies must be tunable so that the difference frequency $\omega_1 - \omega_2$ can be matched with the resonance of interest. Pulse energies greater than 100 μJ are required to study the dynamics of highly excited materials. To probe these dynamics on a subpicosecond time scale, pulse widths must be on the order of 100 fs or less. Finally, because all of these techniques involve the overlap of two or more beams, a smooth spatial beam profile is essential. Thus, progress in nonlinear laser spectroscopy and ultrafast phenomena will be greatly enhanced by laser systems with the following six characteristics: (i) two beams at different frequencies, (ii) tunability, (iii) time synchronization, (iv) high pulse energy, (v) femtosecond pulse width, and (vi) excellent spatial profile.

Previous attempts at such a laser system suffer from one of two main problems: time jitter and poor spatial profile. The synchronously pumped dye laser system can produce femtosecond pulses, but the time jitter between the two beams is a few picoseconds, severely limiting the temporal resolution [1,2]. The synchronization problem can be solved by using part of one beam to generate a continuum in a nonlinear medium which then becomes the source for the second beam [3–5]. However, this method is accompanied generally by self-focusing in the nonlinear medium which leads to the spatial breakup of the beam into random light filaments [#1] [6].

We use continuum generation in a single-mode, polarization-preserving fiber to solve the synchronization problem while completely avoiding self-focusing effects [7]. The output of our fiber is amplified in a Bethune cell dye amplifier which preserves the nearly perfect gaussian spatial profile of the fiber output while achieving a final output energy of 100–

[#1] To generate high energy pulses, we use a low repetition rate system. Although in a high repetition rate system the input energy for white light generation can be controlled such that it is above the threshold for self-phase modulation but below that for self-focusing, the same does not hold for low repetition rate systems. The energy fluctuations of low repetition rate systems make it impossible to maintain the input energy within the narrow energy window between the threshold for self-phase modulation and that for self-focusing. For details on self-focusing and self-phase modulation, see ref. [6].

0030-4018/92/$05.00

200 μJ [8]. Efficient amplification results from temporal pulse broadening in the fiber due to positive group velocity dispersion [2] [9]. The broadened spectrum and linear frequency chirp of the amplified output allow temporal pulse compression to a pulse width shorter than that of the input to the fiber [10–12]. Our pulses are compressed by a grating pair to 42 fs (fwhm).

Although single-mode fibers are used in other ultrafast laser systems for continuum generation and pulse compression purposes, none of these systems exhibits all six of the above characteristics [13]. The combination of the single-mode fiber and the Bethune cell amplifier is a novel and potentially very useful to designing a laser system particularly well suited to the fields of nonlinear laser spectroscopy and ultrafast phenomena.

Fig. 1 shows a schematic diagram of the fiber-amplifier setup. An amplified colliding-pulse-mode-locked (CPM) system [3] [14] pumped with a 10 Hz, 300 mJ, frequency-doubled *Q*-switched Nd:YAG laser produces 600 μJ, 150 fs (fwhm) pulses centered at 620 nm. Most of this output is used without further modification as a pump beam for pump-probe experiments or split into two ω_1 beams for four-wave mixing techniques. The remainder of the energy is used in a second beam to generate a pulse train at a tunable frequency ω_2 that is synchronized with the ω_1 beam.

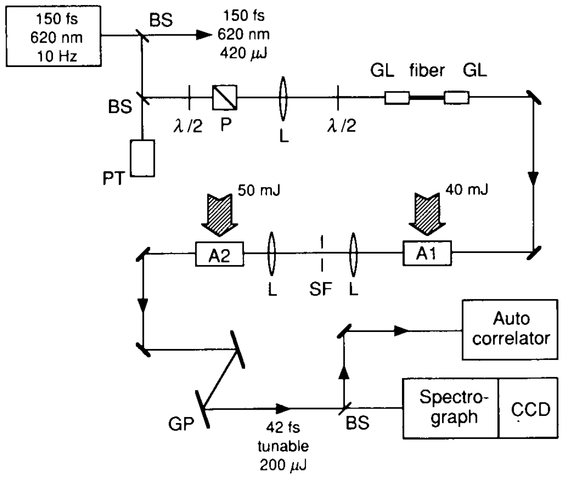

Fig. 1. Schematic diagram of the fiber-amplifier setup. BS, beam splitter; λ/2, broadband half-wave plate; P, polarizer; PT, phototube; GL, gradient index lens; A1, first amplifier stage; L, lens, SF, spatial filter; A2, second amplifier stage; GP, grating pair.

This second beam is sent into a 20 mm, single-mode, polarization-preserving fiber with a 3.3 μm core diameter to provide a supercontinuum. To prevent damaging the fiber, a half-wave plate and a polarizer cube are used to limit the input energy to approximately 1.5 μJ [4]. The polarization of the laser beam is matched to the birefringent axis of the fiber with another half-wave plate, and two gradient index rods couple the beam into and out of the fiber.

The output from the fiber is amplified in two Bethune prism dye cells, which preserve the Gaussian spatial profile. To amplify the long-wavelength region of the fiber output we use DCM dissolved in methanol with the dye concentration optimized for uniform pumping [5] [15]. The first dye cell has a 2 mm bore diameter and is pumped with 40 mJ laser pulses from the same Nd:YAG laser that pumps the main amplifier. The second cell, with a 3 mm bore diameter, is pumped with 50 mJ pulses produced by doubling the residual infrared energy from the Nd:YAG laser. A spatial filter with an 80 μm pinhole between the two dye cells suppresses amplified spontaneous emission. The amplified pulses are recompressed temporally by two 600 grooves/mm diffraction gratings, which provide negative group velocity dispersion to counter the positive dispersion in the fiber-amplifier setup [16].

The amplified output has a broad spectral range and high energy, and exhibits a shorter pulsewidth and cleaner spatial profile than the input to the fiber. The fiber produces a useful spectral range greater than 200 nm, compared to the 5 nm bandwidth of the input pulses. The single-shot spectrum in fig. 2 cor-

---

[2] This effect is similar to the chirped pulse amplification discussed in ref. [9].

[3] Our amplifier is similar to the one described in ref. [14].

[4] Our coupling efficiency into the fiber is less than 10% because the input beam cannot be focused tightly enough, so most of the 1.5 μJ is lost at the front face of the fiber. However, the relatively large size of the input beam allows for easier alignment, so the low coupling efficiency is actually a practical advantage.

[5] Uniform pumping of a dye solution with an absorption coefficient α at the pump frequency in a Bethune prism cell with hole radius r occurs for $\alpha r = 0.75$. See ref. [15].

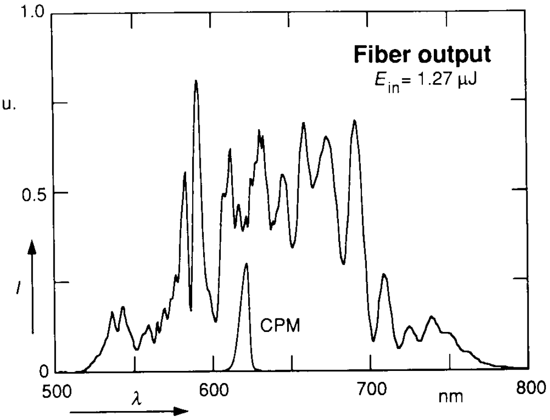

Fig. 2. Single-shot spectrum of the fiber output obtained at an input energy of 1.27 μJ. The narrow spectrum centered at 620 nm is that of the output of the CPM.

responds to a fiber input energy of 1.27 μJ, which is about 13 times the threshold for self-phase modulation in the fiber. We measured the input energy dependence of the spectral broadening and found that the width of the broadened spectrum is independent of input energy above a critical value of 10 times the threshold. Thus, shot-to-shot stability in the bandwidth of the spectrum is assured by maintaining the input energy above this critical value. Following amplification, the bandwidth is determined by the spectral range of the amplifying dye (cf. figs. 2 and 3). Note that the single-shot amplified pulse spectrum shown in fig. 3 exhibits significantly less structure than the single-shot unamplified pulse spectrum in fig. 2. This spectral smoothening is due largely to gain saturation in the amplifier, which also gives shot-to-shot spectral and energy stability.

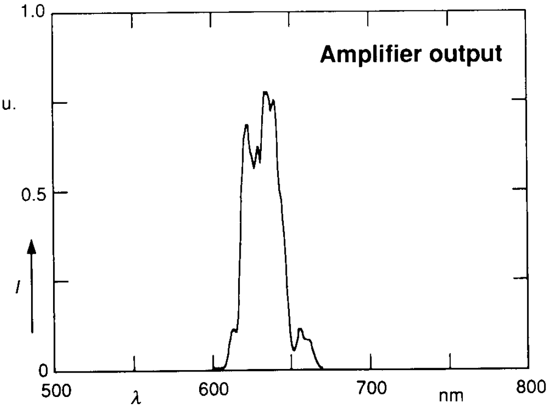

Fig. 3. Single-shot spectrum of the output from the two-stage amplifier using DCM dissolved in methanol. The width of the spectrum reflects the gain–bandwidth of the dye.

The amplifier after the fiber has a first-stage gain of 5000 and a second stage gain of 40, yielding a total energy of 100–200 μJ, less than 1% of which is amplified spontaneous emission. This efficient amplification is possible because of the spectral and temporal broadening of the pulse by the fiber. In addition, the broadened spectrum and the linear frequency chirp allow compression of the pulse to a shorter duration than that of the input pulse to the fiber. With a perpendicular distance of 55 mm separating the two gratings, we obtain a single-shot pulsewidth of 42 fs (fwhm); a deviation of 5 mm in the grating separation distance results in a 35% increase in the pulsewidth. The main benefit of Bethune cell amplification of the output from a single-mode fiber is the smooth and stable spatial profile that results. Fig. 4 shows the far-field pattern of the amplified beam photographed using a CCD camera at a distance of 3 m from the last amplifier stage, as well as profiles along the x- and y-axes which are almost perfect gaussians. Moreover, the spatial profile shows very little shot-to-shot fluctuation, which is a distinct advantage for techniques involving beam overlap.

The flexibility of the system can be increased by placing the fiber immediately after a one- or two-stage pre-amplifier rather than splitting off the input to the fiber from a high-energy beam. Then, a beamsplitter can be placed after the fiber resulting in two super-continuum beams, each sent through a separate dye amplifier to provide two broadband, high-energy pulse trains at different wavelengths. About 350 mJ of frequency-doubled Nd:YAG pulse energy should be sufficient to pump the pre-amplifier as well as a three-stage amplifier and a two-stage amplifier following the fiber with respective outputs of 1 mJ and 200 μJ per pulse. We are currently implementing such a design. Both beams will then be independently tunable and compressible to 40–50 fs. Furthermore, the nearly perfect spatial profile of the two beams should greatly reduce experimental noise caused by fluctuations in beam overlap at the sample. With a broader gain-bandwidth amplifier, such as a Ti:sapphire-

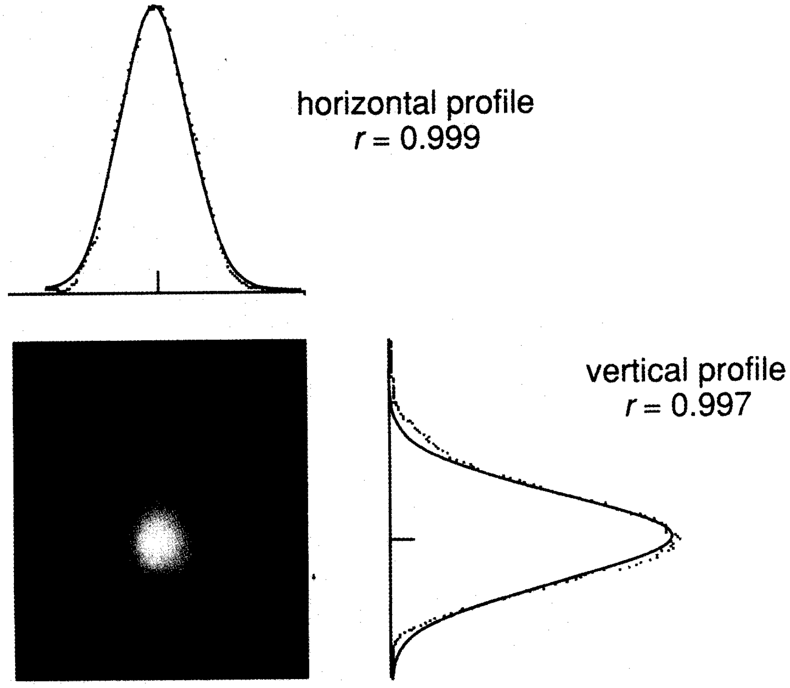

Fig. 4. Single-shot far-field spatial profile of the amplified pulse. The image was taken using a CCD camera. The dots in the horizontal and vertical profiles are data points; the solid lines are gaussian fits; r is the correlation coefficient of the fit.

based amplifier, this technique could produce even shorter pulsewidths while maintaining high energies.

In conclusion, we present a versatile femtosecond laser system producing two beams at different frequencies and synchronized in time. Tunability, high energy and excellent spatial profile are provided by the combination of supercontinuum generation in a single-mode fiber and amplification in a two-stage Bethune cell amplifier. The output from this fiber-amplifier combination, which is compressed by a grating pair to 42 fs pulse width (fwhm), has a tuning range greater than 200 nm and pulse energies of 100–200 μJ. Because the shortcomings of time jitter and poor spatial profile are eliminated, this system is particularly well suited for high-energy dual-wavelength pump-probe experiments and ultrafast nonlinear laser spectroscopy.

Acknowledgements

The authors thank Jay Goldman, Peter Saeta, and Eli Glezer for many useful comments. This research is supported by National Science Foundation grant DMR-8920490 and the Joint Services Electronics Program under contract N00014-89-J-1023. Y.S. gratefully acknowledges the support of a National Science Foundation Graduate Fellowship.

References

[1] W. Zinth, R. Leonhardt, W. Holzapfel and W. Kaiser, IEEE J. Quantum Electron. QE-24 (1988) 455.

[2] T. Juhasz, G.O. Smith, S.M. Mehta, K. Harris and W.E. Bron, IEEE J. Quantum Electron. QE-25 (1989) 1704.

[3] A. Migus, A. Antonetti, K. Etchepare, D. Hulin and A. Orszag, J. Opt. Soc. Am. B 2 (1985) 584.

[4] J.H. Glownia, J. Misewich and P.P. Sorokin, in: The supercontinuum laser source, ed. R.R. Alfano (Springer, Berlin, 1989) pp. 337–376.

[5] N.D. Hung and Y.H. Meyer, Appl. Phys. B 52 (1991) 67.

[6] Y.R. Shen and G.-Z. Yang, in: The supercontinuum laser source, ed. R.R. Alfano (Springer, Berlin, 1989) pp. 1–32; I. Golub, Optics Lett. 15 (1990) 305.

[7] G.P. Agrawal, Nonlinear fiber optics (Academic Press, New York, 1989) ch. 4.

[8] D.S. Bethune, Appl. Optics 20 (1981) 1897.

[9] P. Maine, D. Strickland, P. Bado, M. Pessot and G. Mourou, IEEE J. Quantum Electron. QE-24 (1988) 398.

[10] C.V. Shank, R.L. Fork, R. Yen, R.H. Stolen and W.J. Tomlinson, Appl. Phys. Lett. 40 (1982) 761.

[11] D. Grischkowsky and A.C. Balant, Appl. Phys. Lett. 41 (1982) 1.

[12] R.L. Fork, C.H.B. Cruz, P.C. Becker and C.V. Shank, Optics Lett. 12 (1987) 483.

[13] G. Boyer, M. Franco, J.P. Chambaret, A. Migus, A. Antonetti, P. Georges, F. Salin and A. Brun, Appl. Phys. Lett. 53 (1988) 823.

[14] M.M. Murnane and R.W. Falcone, J. Opt. Soc. Am. B 5 (1988) 1573.

[15] F.P. Schäfer, Appl. Phys. B 39 (1986) 1.

[16] E.B. Treacy, IEEE J. Quantum Electron. QE-5 (1969) 454.