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Generation of 0.6 μJ pulses of 16 fs duration through high-repetition rate amplification of self-phase modulated pulses

G. Boyer, M. Franco, J. P. Chambaret, A. Migus, and A. Antonetti
Laboratoire d'Optique Appliquée, ENSTA-Ecole Polytechnique, 91120 Palaiseau, France

P. Georges, F. Salin, and A. Brun
Institut d'Optique Théorique et Appliquée, Université Paris-Sud, 91406 Orsay Cedex, France

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We have generated optical pulses of 16 fs duration in the submicrojoule range and at a repetition rate of 11 kHz. This was obtained by amplifying self-phase modulated pulses at the output of a fiber, using a couple of dyes to achieve gain covering a large spectrum around 620 nm.

In this letter we report a new scheme combining different stages of amplification and compression, producing, at a repetition rate of 11 kHz, pulses in the microjoule range with durations as short as 16 fs around 620 nm. We start from the particularly successful approach to the generation of ultrashort pulses based on the principle of self-phase modulation (SPM) in an optical fiber and further compression, a technique which has been recently optimized to produce pulses as short as 6 fs.¹ In this type of experiment, optical pulses of a few tens of femtoseconds are first amplified and next focused into a single-mode fiber a few millimeters long, where they undergo group velocity dispersion and self-phase modulation. The net effect of the combination of these two effects is a spectral broadening associated with a quasi-linear chirp of the pulse.² Compensation of this chirp through a negative delay line (grating pair, four-prism arrangement or combination of both) finally yields compression down to a few femtoseconds. While these recent achievements have found immediate applications in time-resolved spectroscopy,³ the limitation of this method lies in the damage threshold level of the single-mode fiber (about 3 TW/cm²) which allows the production of pulses of only a few nanojoules in energy. Recently, Knox reported the amplification of compressed pulses to an unspecified energy, but was limited to 26 fs duration due to the available gain bandwidth.⁴ Even more recently, Rolland and Corkum have been able to compress high-power pulses in bulk material and then obtain 100 μJ of energy for 24 fs pulses, reduced to 7 μJ of available energy for 19 fs pulses,⁵ but at a much smaller repetition rate than the one reported here.

The basic idea we have applied in the course of this work was to amplify the self-phase modulated and chirped pulse obtained at the fiber output, using a broadband amplifying medium.⁶ This technique has some similarities with the "chirped pulses amplification" developed by Strickland and Mourou⁷ for Nd-glass lasers, but we do not expand the pulse duration to a large value. In our case the principal obstacle lies in the spectrum width of the pulses to amplify, (over 50 nm for a 10 fs duration pulse in the visible). A capability of gain covering such a broadband requires either a mixture of known gain media or new materials. We have worked out the first solution using different mixtures of dyes. The main difficulty to overcome in this approach is the Förster electronic energy transfer which occurs with high efficiency in case two

molecules undergo dipole allowed transition and the emission spectrum of the donor molecule overlaps well the absorption spectrum of the acceptor.⁸ This means we have to couple dyes with the constraints that the emission band of one does not fall within the absorption of the other but with a gain overlap covering the desired spectrum without any interruption. Such a couple, DCM (4-dicyanomethylene-2-methyl-6p-dimethylaminostyryl-4Hpyran) and rhodamine 610, has been recently shown⁶ to be suitable for amplifying a large spectrum around 620 nm since the DCM peak absorption (480 nm) is well outside the emission band of Rh610 (peaked at 588 nm), while both dyes absorb almost equally well the green 511 nm line of the copper vapor laser and

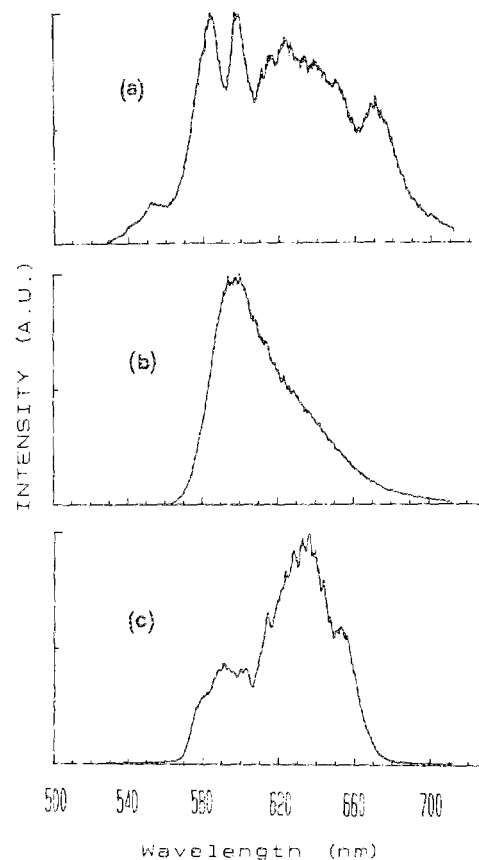


FIG. 1. Spectrum of the pulse at the fiber output (a), after amplification (c), and fluorescence of the dye mixture (b).

generate together an emission spectrum covering the region of interest [Fig. 1(b)].

The initial part of the setup is very similar to the one developed by Knox *et al.*⁹ It includes a colliding pulse mode-locked (CPM) dye ring laser with an intracavity four-prism arrangement, producing two output beams at 620 nm, consisting of pulses delivered at a rate of 100 MHz, of duration 40 fs and energy 0.1 nJ. One of the two output beams undergoes a five-pass amplification in a Rh640 ethylene glycol jet pumped by half of the 15 W of the green line (511 nm) of a copper vapor laser (CVL) running at 11 kHz (Cu40 from Oxford Lasers, delivering 40 W all lines, using an off-axis unstable cavity). Group velocity dispersion in the amplifiers and the optics is compensated using a four-(high index) prism¹⁰ arrangement so that output pulses are typically 50 fs long and have an energy of 2 μ J.

A small part (50 nJ at maximum) of this energy is then coupled into a 11-mm-long (3- μ m diameter) standard single-mode fiber, inducing spectral broadening and group velocity dispersion so that at the output the chirped pulses have an energy in the range 3–4 nJ, a duration of 300 fs, and a spectrum extending over 80 nm or more [see Fig. 1(a)]. This beam is then sent into a five-pass amplifier similar to the previous one (Fig. 2). The gain medium, a mixture of DCM (concentration 2×10^{-4} M/l) and Rh610 (concentration 3×10^{-4} M/l) dissolved in a mixture of ethylene glycol and propylene carbonate in the ratio 3 to 1, is flowing through a sapphire nozzle 1.25 mm thick. The pumping source is the other half (above 7 W) of the CVL line at 511 nm, an arrangement which readily yields time synchronization with the amplified pulse. The gain in energy after five passes reaches a factor above 300, corresponding therefore to an energy of 1 μ J per pulse. Surprisingly, the spectrum after amplification [Fig. 1(c)] is nearly as broad as the input one with, however, a small hole around 600 nm. No saturable absorber was needed since the amplified spontaneous emission had a very small contribution (less than 1%) in the

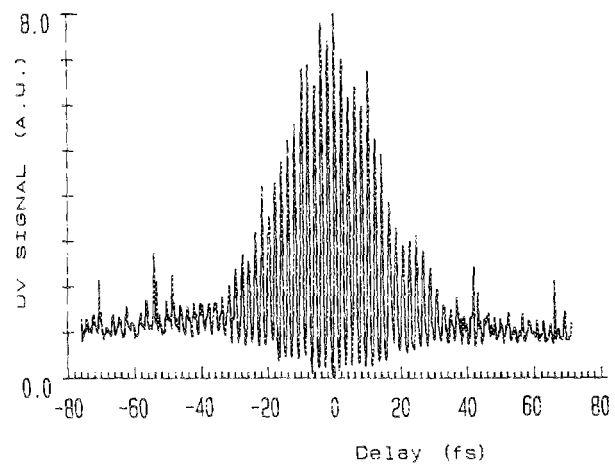


FIG. 3. Interferometric autocorrelation of the 16 fs amplified pulse.

output energy. However, we should point out that a good focusing of the femtosecond beam in the amplifier was crucial for getting such a large gain. This was obtained through the use of five lenses, ensuring a beam diameter expanding from about 30 μ m in the first pass to 300 μ m in the last one. The gain per pass, slightly above three, is smaller than in the first amplifier, which is due to the smaller emission cross section of these infrared dyes. This means that achievement of the same total gain will need more passes. Note that this does not affect the available amplified energy (1 μ J in the second amplifier versus 2 μ J in the first one).

Finally, the amplified beam traveled through a sequence of four high-index glass prisms (Schott SF10 glass) in view of compensating the group velocity dispersion in the initial fiber, in the amplifying medium, and in the optics. Due to the fact that the fiber we used did not preserve the polarization, losses in the prism sequence were not negligible, so that the output energy was 0.6 μ J at maximum. The amplified pulse duration was analyzed following the interferometric autocorrelation scheme, using a 0.1-mm-thick KDP crystal. Figure 3 represents such an autocorrelation which fits relatively

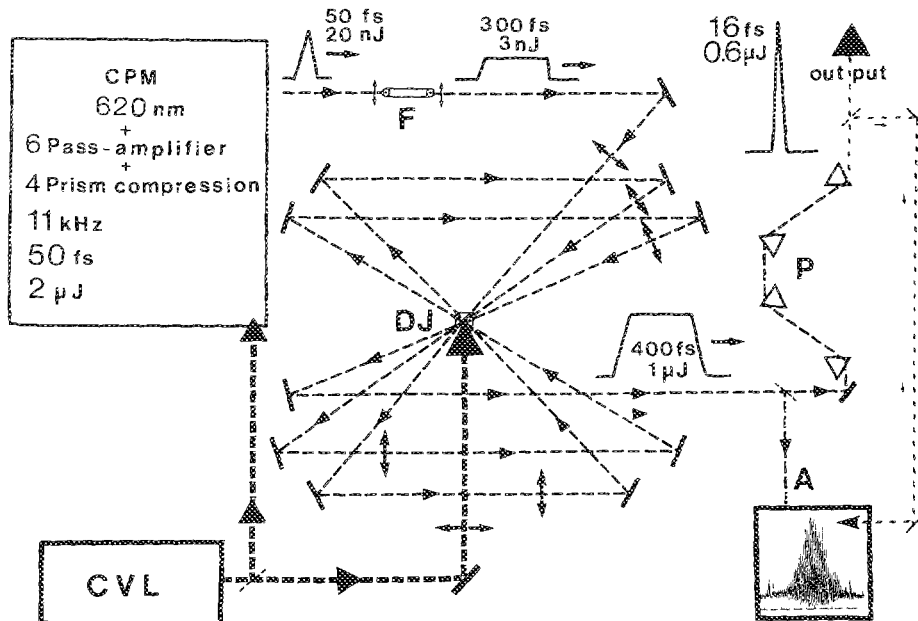


FIG. 2. Sketch of the femtosecond broadband amplifier. After initial amplification part of the 50 fs pulses are focused into the 11-mm-long fiber (F). At the output the pulses are 300 fs long but have a bandwidth above 50 nm. They next undergo a five-pass amplification in the dye jet (DJ) composed of a mixture of Rh640 and DCM, up to an energy of 1 μ J. They are finally compressed to 16 fs in a four-prism arrangement.

well with a hyperbolic secant square pulse of duration 16 fs. We notice, however, that the output spectrum width (Fig. 1) is able to sustain a pulse of much shorter duration, which means that phase higher order terms are not compensated correctly. This is not surprising since it is known that third-order terms are important in high-index prisms. This was verified by replacing the prisms by a two-pass grating pair with which we could obtain less than 12 fs pulses but at the expense of much greater losses. We want also to point out that the quality of the mode was good enough to generate a spectral continuum by focusing less than $0.5 \mu\text{J}$ of the 16 fs pulses into a 1-mm-thick glass plate. This implies that time-resolved pump *and* probe experiments are now possible on this time scale in a spectral range extending from the near ultraviolet to the near infrared.

We now turn to the principle of the experiment, namely, the amplification of a broadband spectrum. We found it quite surprising to get such good results since we expected that gain narrowing¹¹ should play an essential role in limiting the bandwidth and, therefore, the pulse duration.⁴ It turns out that the fluorescence curve shown in Fig. 2 associated to a gain of 300 should not allow the generation of the amplified spectrum [Fig. 2(c)]. This suggests that nonlinear effects such as gain saturation leading to phase modulation, occur in the dye jet, compensating or even overcompensating, the gain narrowing. Side lobes should then be generated on both sides around the main amplified peak, but absorption of rhodamine 610 prevents any emission under 570 nm, which explains the sharp high-energy side of the resulting spectrum. We should, however, point out that the combined effects of phase modulation, and selective gain or absorption did not destroy the quasi-linear chirp in the pulse.

In conclusion, we have amplified self-phase modulated pulses to the submicrojoule level while keeping a duration shorter than 16 fs. This system, operating at a 11 kHz repetition rate, allows already time-resolved spectroscopy experiments on this time scale, but we anticipate that pulses as short as 10 fs should be obtained in this energy range using a better compensation of phase distortions.

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