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Patrick Georges, François Salin, Gilles Le Saux, Gérard Roger, Alain Brun. 20 fs amplified pulses. Optics Communications, 1988, 67 (4), pp.297-300. 10.1016/0030-4018(88)90153-8 . hal-00709927

HAL Id: hal-00709927

<https://hal-iogs.archives-ouvertes.fr/hal-00709927>

Submitted on 8 Sep 2022

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20 fs AMPLIFIED PULSES

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We describe the production of 20 fs, a few microjoule pulses using amplification and compression of 80 fs pulses spectrally broadened by self-phase modulation in an optical fiber.

With the development of mode-locked lasers, the temporal resolution of physical phenomena kinetics measurements is limited by the laser pulse width. The compression of optical pulses spectrally broadened by self-phase modulation [1] is the method generally used to obtain the shortest pulses [2]. Recently, Fork et al. have reported the compression of optical pulses to six femtoseconds [3]. But unfortunately, the single mode fibers used limit the input power to a few hundred kilowatts. Moreover, the study of nonlinear interactions induced by optical pulses needs high energy levels. These compressed pulses can not be used as a pump in such experiments. At this time, the temporal resolution is limited around 50 fs [4] by the duration of the high peak power amplified pulses produced by the classical system containing a colliding pulse mode locked laser (CPM) and an

amplifier. In this paper, we report the generation of 20 fs pulses with energy of few microjoules. The principle is the following: an optical pulse is spectrally broadened by self-phase modulation in a single mode fiber; the output pulse is amplified in a multipass amplifier and then compressed by a sequence of four prisms. The obtained energy level is sufficient to generate nonlinear phenomena such as spectral continuum.

In our experiment, we have used optical pulses produced by a CPM laser and amplified with a 3 ns Nd:YAG laser at a 10 Hz repetition rate to energies of about 1 mJ. The pulse duration is 80 fs and their spectrum is centered at 615 nm. After a filtering to improve the spatial profile of the beam, a fraction of the pulse energy is coupled with a 20× microscope objective into a 15 mm long single mode fiber with

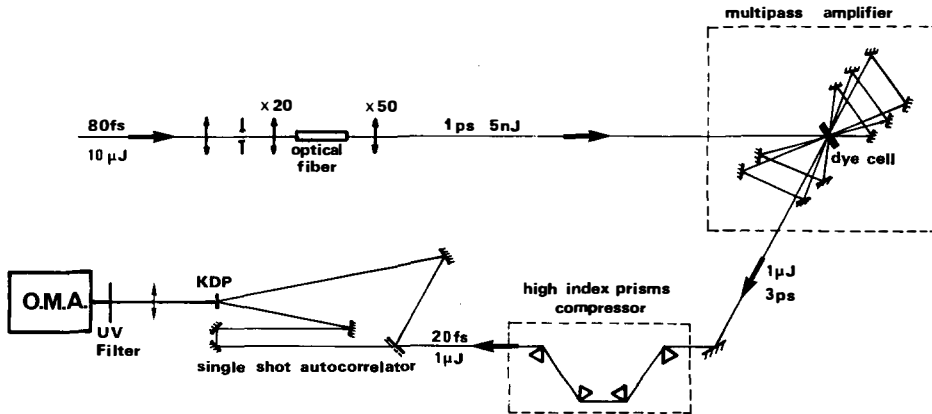


Fig. 1. Experimental set-up.

a 4 μm core diameter (fig. 1). Looking at the output pulse spectrum, we estimate that the intensity in the fiber is close to 50 kW, which is well below the damage threshold of the fiber input face. A typical pulse spectrum at the output of the fiber is shown in fig. 2. One can see an asymmetrical shift of the spectrum towards the shorter wavelengths. Similar spectra have already been observed by Knox et al. [5] and studied by Bourkoff et al. [6] taking high order nonlinearity and dispersion terms into account. At the fiber output, the pulse has been lengthened by group velocity dispersion and its duration is estimated to about 1 ps. The beam is then collimated with a 50 \times microscope objective. To increase the energy, we amplify the pulse before compression. In fact, long pulses make the amplification more efficient and limit nonlinear effects in this process. Furthermore, recent experiments using solid state amplifiers and linearly chirped pulses [7] have shown that the phase relation between the frequency components is conserved after the amplification allowing good compression ratios.

We use a multipass amplifier [8] to improve the gain efficiency. The pulse passes six times through a 5 mm dye cell pumped by a 20 mJ, 3 ns pulse produced by a Nd:YAG laser (fig. 1). The main problem is the choice of the amplifier dye. This dye must have a good efficiency in a large spectral band centered on the laser wavelength. In fact, the com-

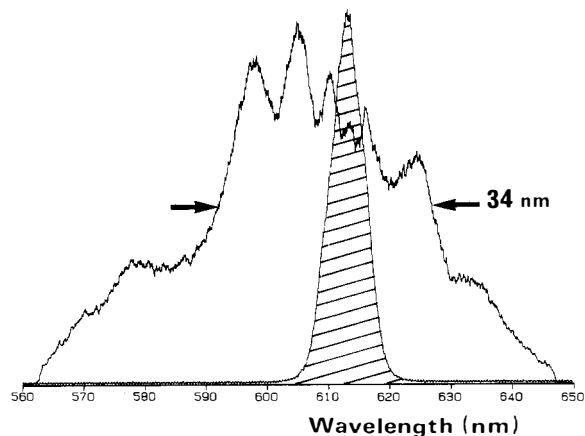


Fig. 2. Broadened pulse spectrum at the output of the fiber. The input pulse peak power is estimated from the spectrum fwhm at 50 kW. The input spectrum is given as a comparison (hatched curve).

pressed pulse duration will be limited by the possibility of amplifying a large spectral band, where the chirp of the pulse is linear. We have studied several dyes to amplify the large output spectrum issued from the fiber [9], but either they had not a sufficient spectral bandwidth (Sulforhodamine 640, Rhodamine 640) or their bandwidths were large enough but not centered on the input laser spectrum (Rhodamine 610, Kiton Red). So we decided to mix two dyes to center and increase the bandwidth of the amplified spectrum. The problem is that the fluorescence of the first dye must not be absorbed by the second dye. Two dyes: Rhodamine 610 and DCM (4 dicyanomethylene-2-methyl-6p-dimethylaminos-tyrlyl-4H-pyran) were used because they have complementary spectral bandwidths and because the DCM absorption maximum (481 nm) is at a lower wavelength than the Rhodamine 610 fluorescence maximum (588 nm). Dye concentrations were approximately 2×10^{-5} M/l for the DCM and 2×10^{-5} M/l for the Rhodamine 610 (dissolved in methanol). Fig. 3 shows a typical amplified spectrum with a mixture of Rhodamine 610 and DCM. The full width at half maximum (fwhm) is 30 nm. The asymmetry of the amplified spectrum could come from an imperfect relative concentration of the two dyes. We are now studying other dye concentrations and mixtures in order to increase the amplified bandwidth. We have measured an efficiency gain for our multipass amplifier close to one thousand with such a large amplification bandwidth. Thus, we es-

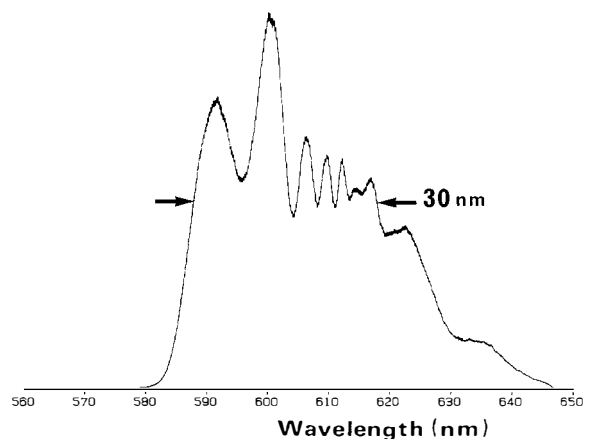


Fig. 3. Amplified pulse spectrum using a DCM (2×10^{-5} M/l) and Rhodamine 610 (2×10^{-5} M/l) mixture.

timate the amplified pulse energy to about a few microjoules.

The amplified chirped pulses are then compressed with a sequence of four high index refraction prisms (Fed 0525 from Sovirel) [10]. The use of prisms offers many advantages: ease in alignment and in adjustment (much less critical than with gratings) and very high transmission ratio. The prisms have an apex distance of about 28 cm and compensate the dispersion produced both in the fiber and in the amplifier.

The compressed optical pulses are measured using a single shot correlator [11]. With an optical multichannel analyzer (O.M.A.), we record the spatial profile of the second harmonic beam produced when two expanded beams cross in a non-linear crystal 0.3 mm (KDP). This system gives a real time visualization of the pulse autocorrelation and then allows us a precise adjustment of the prisms compressor. We regularly obtain single-shot autocorrelation functions, which correspond to pulses fwhm under 25 fs (assuming a hyperbolic secant pulse shape). The shortest pulses obtained (fig. 4) have a fwhm of 20 fs. The corresponding spectral width as seen from fig. 3 is 30 nm. For this spectral width (and a hyperbolic secant pulse shape), a minimum pulse width of 13 fs is possible, indicating that our pulses are close to the Fourier transform limit. One can see that the autocorrelation function presents wings. We think, that it corresponds to a nonlinear chirp which is not compensated by our compressor system [12].

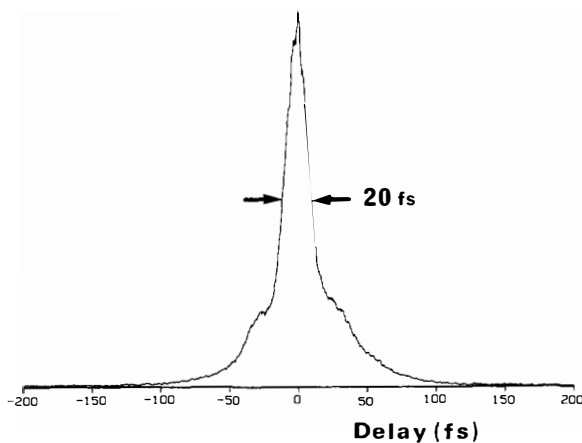


Fig. 4. Single shot autocorrelation of a 20 fs (50 MW) amplified and compressed pulse.

In fact, if the sequence of four high index refraction prisms is very compact and have a good transmission ratio, it presents a large negative cubic dispersion. The fiber positive cubic dispersion is thus overcompensated. We are presently studying a compressor system containing both two gratings and two prisms [2] to resolve this problem and to obtain pulses close to Fourier transform limitation.

The stability of the compressed pulses directly depends on the spectral broadening in the optical fiber. The main instability sources are the incident femtosecond pulses fluctuations and the variations in the fiber coupling. By averaging twenty consecutive autocorrelations traces, we observe a broadening from 20 to 24 fs (fig. 5). This small broadening indicates a good shot to shot stability. Moreover we have also measured the beam divergence using a long focal lens (2 m) and a detector array. Results show the 20 fs pulses to be 1.1 times diffraction limited. Therefore it seems that the amplifier does not affect the spatial beam profile. This beam once focussed could produce intensity in the range of 10^{15} W/cm². To prove the possibility of using these amplified ultrashort pulses in spectroscopic experiments, we have focused the beam in a 1 mm cell of water using a 35 mm lens. We then observed a spectral continuum extending from UV to IR with a very good pump depletion. The stability of the continuum was comparable to the one obtained using the initial 80 fs pulses.

In this paper, we present the generation of high energy 20 fs pulses. The broad spectrum produced in

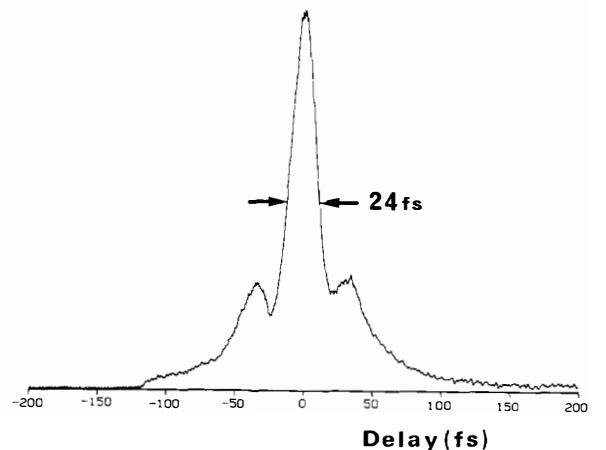


Fig. 5. Twenty-shots averaged autocorrelation function.

a single mode fiber by self-phase modulation is amplified in a multipass amplifier using a mixture of two dyes. The pulse energy is sufficient to create nonlinear effects such as a continuum and so decrease our temporal resolution in spectroscopic experiments under 20 fs. To our knowledge these are the shortest amplified pulses produced at this time.

We thank H. Severac and J.L. Tapie for valuable technical assistance for the multipass amplifier realization. This work is supported in part by the Ministère de la Recherche Scientifique and by the Direction des recherches et Etudes Techniques (Division Optique).

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