Quantum Electronics **42** (12) 1097 – 1099 (2012)

PACS numbers: 42.65.Re; 42.65.Jx; 42.65.Ky
DOI: 10.1070/QE2012v042n12ABEH015067

Spectral broadening and self-compression of negatively chirped visible femtosecond pulses in fused silica

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Abstract. **This paper describes a new effect: spectral broadening and self-compression of negatively chirped visible femtosecond pulses as a result of nonlinear interaction of large-aperture beams with fused silica. We assume that the likely mechanism of the observed spectral broadening is the combined effect of self-phase modulation and four-wave mixing.**

Keywords: *negatively chirped femtosecond pulse, self-compression, self-phase modulation, four-wave mixing.*

The use of the currently most widespread self-compression methods – light filamentation in gases (see e.g. Refs [1, 2]) and ionisation-induced self-compression in gas-filled capillary tubes (see e.g. Refs [3, 4]) – is still limited to relatively low pulse energies (from a few millijoules in the former case to a few tens of millijoules in the latter). In connection with this, a search for new mechanisms of femtosecond pulse self-compression, free of the aforementioned drawback, is a critical issue.

Figure 1 shows the experimental configuration we used. Femtosecond pulses (wavelength of ~475 nm) were generated by a frequency-doubled Start 480M Ti:sapphire laser system (Avesta Project Ltd). After spatial filtration and a pulse stretcher based on a prism pair, the beam, with a 1/e diameter of 1 cm and spectral width of 5.1 nm (Fig. 2), was focused by a spherical mirror (R = 1200 mm) onto a sample. The pulse shape and the quadratic phase of the negatively chirped pulse after the stretcher were measured in independent experiments using a SPIDER system (Fig. 3). The sample had the form of a 2.3-mm-thick UV fused silica plate, which was placed 2.5, 3, 4 and 20 cm behind the focal plane. The 1/e beam spot width on the sample surface was then 0.42, 0.50, 0.66 and 3.3 mm, respectively, and the intensity incident on the sample was 1, 0.7, 0.4 and 0.015 TW cm^{-2}. In our experiments, we measured the pulse energy (W = 0.2 mJ), autocorrelation function and spec-

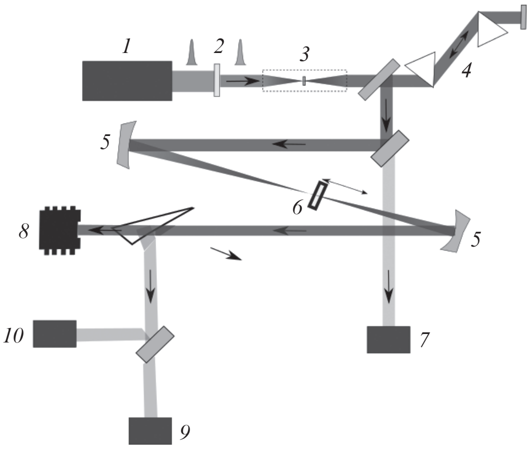

Figure 1. Experimental configuration:
(*1*) Start 480M Ti:sapphire laser system; (*2*) KDP crystal; (*3*) spatial filter; (*4*) prism stretcher; (*5*) spherical mirrors (R = 1200 mm); (*6*) fused silica plate; (*7*) spectrometer; (*8*) calorimeter; (*9*) spectrometer; (*10*) autocorrelator.

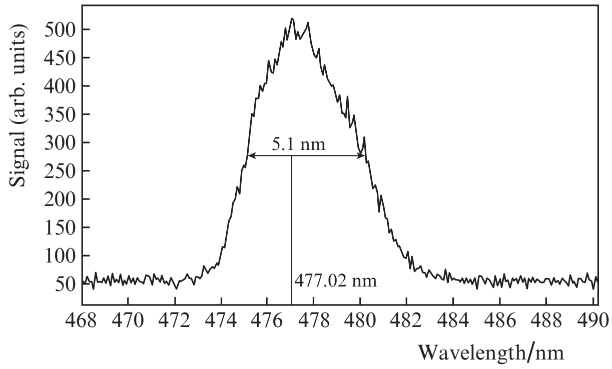

Figure 2. Spectrum of a pulse after the stretcher and before focusing.

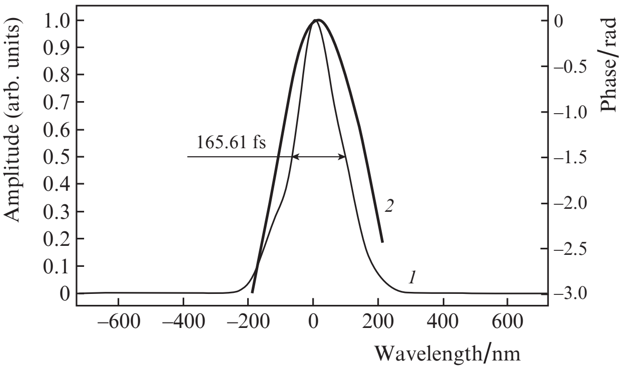

Figure 3. Pulse (*1*) shape and (*2*) phase measured with a SPIDER system behind the stretcher.

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Received 20 November 2012
Kvantovaya Elektronika **42** (12) 1097 – 1099 (2012)
Translated by O.M. Tsarev

trum. The last two characteristics were measured in the central part of the beam, i.e. in the highest intensity region.

The peak laser output power,

$$P = \frac{W}{\sqrt{2\pi}\,\tau_0} = 1.2\ \mathrm{GW}\,,$$

was well below the critical self-focusing power [5, 6],

$$P_{\mathrm{cr}} = \frac{\pi(0.61)^2\lambda^2}{8n_0n_2}(1 + C^2) = 7\ \mathrm{GW},$$

where $C = -2$ is the chirp parameter and $n_2 = 2.4\times10^{-23}$ m^2 W^{-1} is the nonlinear refractive index of air [7]. For this reason, no self-focusing was detected in the beam waist region. At the same time, interaction with air in this region (waist diameter of $\sim$20 μm) led to an additional nonlinear phase shift ($\sim$1.3 rad). This is however of little consequence because, as a result of a phase shift, the effects in question show up at smaller sample thicknesses.

Experimental conditions were adjusted so that, at any intensity, the large-aperture approximation was valid, i.e. the self-focusing distance L_{sf} was considerably greater than the sample thickness. Indeed, since $L_{\mathrm{sf}} = {}^1\!/_2 d[n_0/(2n_2I)]^{1/2}$ [5], where $d = 0.4$ mm is the smallest beam diameter at the highest intensity, $I = 1$ TW cm^{-2}, and $n_2 = 3.3\times10^{-20}$ m^2 W^{-1} [8] is the nonlinear refractive index of fused silica, we obtain $L_{\mathrm{sf}} > 1$ cm.

Figure 4 illustrates the effect of incident intensity, I, on the spectrum and duration of pulses in the sample. Note first of all that the initial spectral shape of the pulse remained almost unchanged behind the focal plane. Its bandwidth (4.8 nm) corresponded to that of a transform-limited Gaussian pulse of 70 fs duration. The initial full width at half maximum of the negatively chirped pulse was 160 fs. It is seen that, for $I < 0.7\times10^{12}$ W cm^{-2}, the spectrum experiences typical narrowing due to self-phase modulation when a negatively chirped pulse propagates through a positive group velocity dispersion medium, and the pulse duration decreases slightly owing to the compensation of the quadratic phase introduced by the prism pair. At higher incident intensities, the spectrum has a more complex structure and the beam experiences time-domain self-compression to a pulse duration of 40–50 fs, which is considerably shorter than the duration of a transform-limited pulse corresponding to the initial width of the spectrum. With increasing incident intensity, the spacing between the peaks in the spectrum decreases. In particular, at intensities of 0.4, 0.7 and 1.0 TW cm^{-2}, the spacing between the peaks is 4.7, 3.8 and 2.7 nm, respectively.

One possible mechanism of the observed effect is the combined influence of self-phase modulation and four-wave mixing. Note that self-phase modulation influences the time domain phase of the negatively chirped pulse, leading to the formation of a modulated structure of the spectrum, whereas four-wave mixing leads to amplification of the side components. This hypothesis has been taken as a starting point for more detailed experimental and theoretical studies.

Thus, the present results are the first to demonstrate spectral broadening as a result of nonlinear interaction of negatively chirped femtosecond pulses with an optical material having normal dispersion and cubic nonlinearity. In addition, we observed self-compression of femtosecond pulses. These findings open up possibilities for developing a new method for nonlinear self-compression of large-aperture femtosecond beams in optical materials, free of the physical limitations on the pulse energy characteristic of known self-compression processes in filaments and gas-filled capillary tubes. Clearly, such a method would be of practical interest for beams with a rectangular spatial profile.

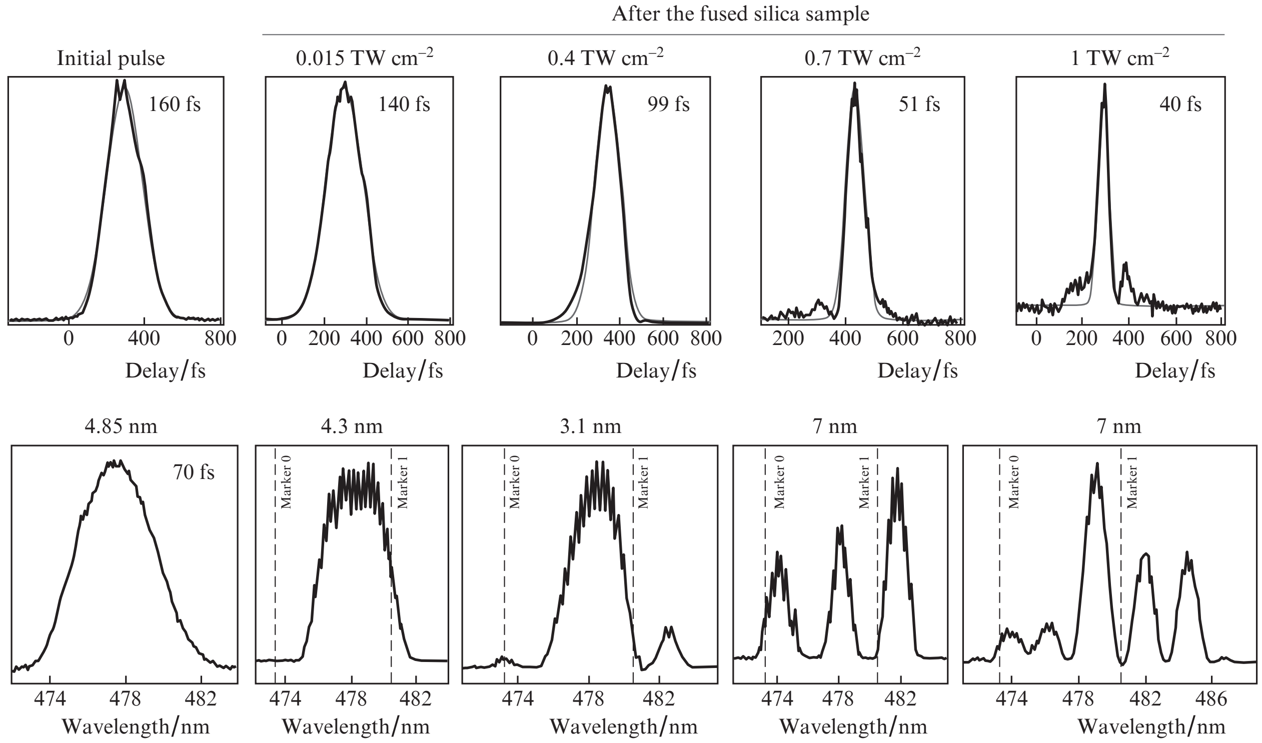

Figure 4. Effect of incident intensity on the autocorrelation traces (top) and spectra (bottom) of pulses before and after the sample.

Self-compression of negatively chirped femtosecond pulses was also observed by Liu et al. [9] when 800-nm light interacted with BK-7 glass. The incident intensity in their experiments was a factor of 1.6 higher than that in this study. They found, however, only a slight narrowing of the spectrum and a change in its shape from Gaussian to Lorentzian, corresponding to a shorter duration of a transform-limited pulse. The discrepancy between their and our results seems to originate from the lower efficiency of nonlinear self-action in the IR spectral region.

Acknowledgements. We are grateful to V.I. Kovalev for useful discussions. This work was supported by the Presidium of the Russian Academy of Sciences (Extremely Strong Optical Fields and Their Applications Programme, Project No. 1.3) and the Russian Foundation for Basic Research (Grant No. 10-02-00121a).

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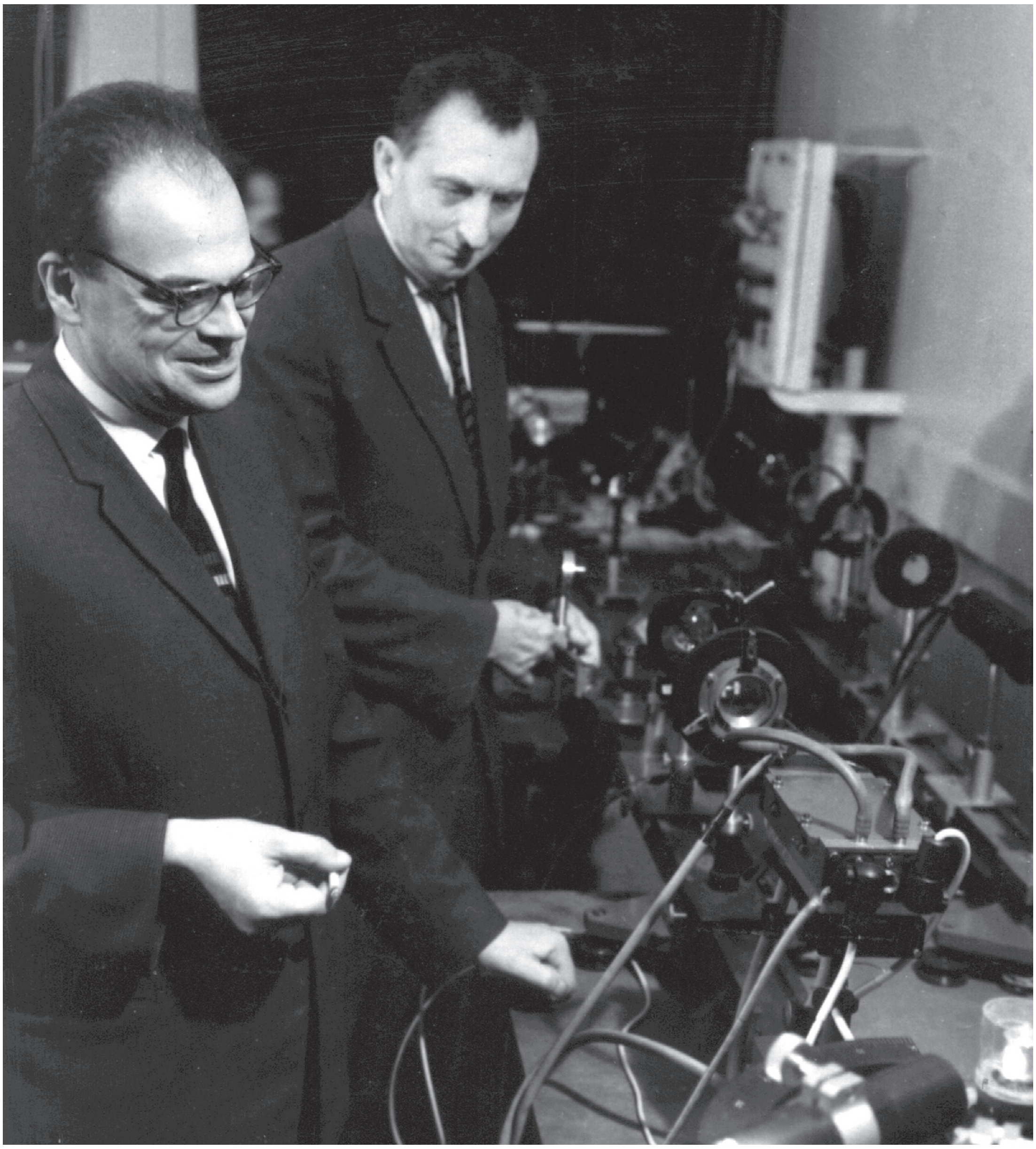

N.G. Basov and A.M. Prokhorov at a laser.