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40-fs hydrogen Raman laser

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Abstract. 40-fs first Stokes pulses at a wavelength of 1.2 μ m were generated in a hydrogen SRS-converter pumped by orthogonally polarised double chirped pulses of a Ti:sapphire laser. To obtain a Stokes pulse close to a transform-limited one, a programmed acousto-optic dispersive delay line was placed between the master oscillator and regenerative amplifier. The energy efficiency of Stokes radiation conversion reached 22%.

Keywords: femtosecond laser systems, dispersive delay line, acoustic diffraction, stimulated Raman scattering.

1. Introduction

In recent years, nonlinear conversion of femtosecond laser pulses aimed at reducing the frequency of initial laser radiation is widely used for obtaining attosecond pulses [1] and high-power terahertz radiation [2]. Efficiency of these processes increases at longer wavelengths of laser radiation. In both the cases, the main reason of higher efficiency is a greater energy of the electron oscillating in a light wave field at a lower frequency of laser radiation.

Crystal parametric generators are mostly often used as nonlinear converters [3]. In this case, the crystal should have a high gain in the corresponding spectral band and a sufficiently broad band of phase matching for femtosecond pulses.

An alternate approach is conversion based on stimulated Raman scattering (SRS). In this case, the band of phase matching with the employment of gas media is substantially greater than in parametric converters on crystals. In addition, Raman converters can be employed for laser pulses with a higher energy. In order to exclude concurrent nonlinear effects such as self-phase modulation and self-focusing which suppress the SRS process, the conversion scheme with a chirped laser pulse duration shorter than hundred picoseconds was suggested [4]. Such pulses are usually obtained with a femtosecond laser system based on amplification of chirped pulses after the amplifier prior to passing into a grating compressor. The converted chirped Stokes pulse is time com-

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Received 21 July 2015 Kvantovaya Elektronika **45** (12) 1101–1104 (2015) Translated by N.A. Raspopov pressed in a grating or prism compressor. This conversion scheme was later improved for eliminating the narrowing of the chirped Stokes pulse spectrum as compared to a laser pulse [5]. Raman conversion in pressurised gases [6] and crystals [7] has been realised by this scheme and Stokes pulses with a spectral width close to that of the initial laser pulse have been obtained.

For maximal compression and minimal duration of compressed Stokes pulse (close to the transform-limited value) to be obtained, it is necessary to compensate for the dispersion of the grating compressor up to the fourth order dispersion. Since the wavelengths of light pulses in the stretcher at the input of a femtosecond laser system amplifier and in the compressor at the output of a Raman converter substantially differ, the high-order dispersion can hardly be compensated for by a simple choice of compressor gratings. In particular, this would require nonstandard gratings with specific numbers of grooves. The problem can also be solved by employing an acousto-optic dispersive delay line (AODDL) for introducing an additional programmed dispersion at the input of the laser amplifier. Actually, this may help to exactly match the dispersion characteristics of the laser amplifier and the Raman converter with those of the grating compressor placed at the system output.

The present work is aimed at possible obtaining minimalduration Stokes pulses in the Raman converter of radiation of a femtosecond Ti:sapphire laser system by using a programmed AODDL.

2. Experimental setup

2.1. Ti:sapphire laser system

An optical scheme of the experimental setup is shown in Fig. 1. The laser system comprises a master oscillator (TiF-15F, Avesta) and a regenerative amplifier (REUS-40F20, Avesta). Between these units, a grating stretcher and a programmed AODDL are placed. Pulses of the master oscillator with the duration of 30 fs and energy of 4 nJ are stretched in time to 100 ps in the grating stretcher, pass through the AODDL and are injected into the cavity of the regenerative amplifier.

The pulse repetition rate of the regenerative amplifier was 20 Hz. The energy of a single chirped pulse with the duration of approximately 100 ps at the output from the regenerative amplifier was 2.5 mJ. The width of the radiation spectrum was about 30 nm with a centre wavelength of 810 nm (Fig. 2), which corresponds to a transform-limited pulse with the duration of 35 fs having a similar spectrum shape. A trans-

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Figure 1. Optical scheme of the experimental setup: (RP) Rochon prism; (FP) film polarisers; (M) mirrors.



Figure 2. Spectra of (a) Ti:sapphire laser output and (b) first Stokes component.

verse intensity distribution of the light beam 3 mm in diameter was close to Gaussian. The beam quality parameter M^2 was at most 1.1.

2.2. Acousto-optic dispersive delay line

The AODDL in the optical channel forms additional dispersions D_1 , D_2 , D_3 and D_4 of appropriate sign and value. The spectral phase is controlled in the quasi-collinear geometry of acousto-optic interaction in a paratellurite single crystal with the angle of ultrasound wave vector inclination of 5° relative to the [110] crystal axis [8, 9]. The dispersive line with the record value of group delay of about 20 ps at a wavelength of 810 nm was used. The geometry of acousto-optic interaction was optimised to make the spectral density of the control high-frequency signal minimal and to increase the diffraction efficiency for ultrashort laser pulses in a wavelength range of 650-880 nm. A scheme of the dispersive delay line is shown in Fig. 3 along with the polarisation directions of input and diffracted radiations. The installation keeps the direction of the axis of the optical system for diffracted spectral components and compensates for the angular dispersion of Bragg synchronism.

An electronic system for controlling the dispersive delay line comprises a digital arbitrary waveform generator with a



Figure 3. Dispersive delay line: (1) paratellurite single crystal; (2) piezoelectric transducer; (3) acoustic absorber; k_i is the wave vector of the zero-order diffraction beam; k_d is the wave vector of the diffracted (delayed/accelerated) spectral component of light; K is the ultrasound wave vector; the incident and diffracted radiations are linearly polarised; the polarisation directions are shown.

sampling rate of 625 million samples per second and a power amplifier. The wave packets generated in a region of acoustooptic interaction are synchronised with a system of laser pulse injection into the regenerative amplifier. The diffraction efficiency at the wavelength of 810 nm in an 80-nm spectral window was above 60% at the peak control power of the high-frequency signal 4 W and at the optical aperture of 4×4 mm. Values of the dispersions arising due to quasi-collinear acoustooptic diffraction of femtosecond radiation on a frequencymodulated ultrasound are determined by the geometry of acousto-optic interaction and are proportional to the length of the interaction region. This AODDL had an extremely long (up to 85 mm) region of interaction with ultrasound as compared to the previously described devices [9-13]. This provided a noticeably wider range of third- and fourth-order dispersions introduced by the delay line. In this case, however, the absolute value of the second-order dispersion produced by the AODDL increases. The value of second-order dispersion D_2 introduced by the AODDL, which is determined by the spectral window width, duration of ultrasound wave packets and coefficient of converting optical frequencies [10] to acoustic, was 4×10^4 fs².

Note that the AODDL introduced into the laser system in front of the regenerative amplifier did not reduce the output pulse energy despite of the AODDL losses. This is explained by the fact that the regenerative amplifier operated in the saturated amplification regime.

2.3. SRS converter and grating compressor

In the Raman converter (Fig. 1) we employed the scheme with double-pulse pumping of the active medium [5]. The optical system comprising a phase half-wave plate $\lambda/2$, two film polarisers FP and two mirrors M, produced two sequenced light pulses with orthogonal linear polarisation. The time interval between the 150-ps chirped pulses was 250 ps. The energy ratio between two pulses was varied by rotating the phase plate. SRS from the first pulse generated the coherent phonon wave in an active medium, on which SRS of the delayed pulse occurred with a high efficiency. The Stokes radiation with the centre wavelength of 1230 nm was separated by a Rochon prism RP.

Hydrogen pressurised to 45 atm was used as an active medium. The length of the cell for Raman conversion was 120 cm. The laser radiation was focused to the cell by a lens with a focal distance of 150 cm.

The time compressor for Stokes radiation was constructed according to the scheme with two gratings. Gold-coated holographic gratings (800 lines mm^{-1}) were used. The energy efficiency of the compressor measured at a wavelength of 1220 nm was 71%.

3. Experimental results and discussion

Prior to carrying out experiments on compression of Stokes radiation, operation of the Raman converter was optimised for obtaining the maximal conversion efficiency at the spectral width of the Stokes radiation pulse close to that of the Ti:sapphire laser. The proportion between energies of two laser pulses with orthogonal polarisations and the hydrogen pressure in the converter cell were varied. It was established that the maximal conversion efficiency of 22% is attained at the energies of the first and delayed pulses 1.2 and 0.9 mJ, respectively. The energy efficiency of the delayed pulse in this case was 50% which corresponds to the photon conversion efficiency of 75%. High conversion efficiency of the delayed chirped pulse favours the fact that the spectral width of Stokes radiation was less than the spectral width of the laser pulse by only 10% (in the frequency scale). In this case, low-frequency spectral components are not cut-off in the spectrum of the Stokes signal (Fig. 2) and the spectral narrowing specific for schemes with pumping by a single positively chirped pulse is not observed [5, 14]. Such a spectrum corresponds to a 39-fs transform-limited pulse.

If the energy of the first pulse increased, the energy redistribution between the pulses resulted in an insignificant spectral broadening of the delayed pulse and a noticeable fall in the energy conversion efficiency. A reduction in the hydrogen pressure also made the efficiency lower. At an elevated pressure, the spatial characteristics of the Stokes light beam became worse, whereas in optimal conditions the intensity distribution was Gaussian.

Calculations performed show that for matching the dispersion characteristics of the compressor with the dispersion introduced by the stretcher and regenerative amplifier, the additional third- and fourth-order dispersions equal to -25×10^4 fs³ and 45×10^4 fs⁴, respectively, should be introduced. The second-order dispersion was compensated for by varying a distance between compressor gratings; the thirdand fourth-order dispersions were compensated for by the AODDL.

Without the acousto-optic delay line the Stokes pulse was compressed down to the duration of about 150 fs, which is approximately four times greater than the duration of a transform-limited pulse with a similar spectrum. In what follows, compression was made by the scheme with the AODDL.

For obtaining the maximal compression (shortest Stokes pulse duration) the values of third- and fourth-order dispersions were varied by the AODDL. One can see from Fig. 4 the duration of the compressed Stokes pulse versus the value of the fourth-order dispersion D_4 at a fixed third-order dispersion D_3 . The pulse duration was measured with an autocorrelator (ASF-20, Avesta) under the assumption that the pulse shape is described by a function sech²t. The shortest pulses of the first Stokes component with the duration of about 38 fs were obtained at $D_3 = -23 \times 10^4$ fs³ and $D_4 = 27.5 \times 10^4$ fs⁴. The autocorrelation function for this pulse is presented in Fig. 5. A noticeable discrepancy between the calculated fourth-order dispersion and the optimal experimental value is related to insufficient accuracy of calculations of this dispersion intro-



Figure 4. Pulse duration τ of Stokes radiation with a centre wavelength of 1220 nm at the output from the grating compressor vs. the dispersion D_4 introduced by the AODDL at $D_3 = -215 \times 10^3$ (\Box), -220×10^3 (\odot), -225×10^3 (\triangle), -230×10^3 (\bigtriangledown) and -235×10^3 fs³(\bullet).



Figure 5. Autocorrelation functions of the radiation pulse at the output from the grating compressor (solid curve) and of the 40-fs pulse described by a function sech²t (dashed curve).

duced by the regenerative amplifier, optical elements of Raman converter and grating compressor.

4. Conclusions

By employing the scheme suggested for Raman conversion of femtosecond laser pulses and introducing the programmed AODDL into the femtosecond laser system we succeeded in obtaining 40-fs pulses of the first Stokes component with an energy of 300 μ J in the case of SRS in hydrogen with the energy conversion efficiency of 22%. Presently, these are the shortest single pulses generated with SRS.

It is worth noting the following:

1. Since SRS conversion in a gas medium of laser pulses with a duration above 100 ps (in the range of intensities substantially below self-focusing and self-phase modulation threshold values) occurs in the saturated regime with the conversion photon efficiency of about 75%, the stability of Raman converter operation with respect to energy and pulse duration is approximately equal to laser operation stability. 2. It is obvious that the pump pulse energy may be increased up to 100 mJ, which is confirmed by SRS conversion of pulses with this energy in gases [15, 16]. In addition, a higher laser pulse energy will require a lower hydrogen pressure for optimal conversion (excess of the pump pulse energy over a threshold SRS value), which, in turn, will increase the time of phase mismatching, reduce wave mismatch and, consequently, make operation of the SRS converter with doublepulse pumping more efficient.

3. A wide spectral transparency range of active gaseous media in Raman converters opens possibilities to assimilate the mid-IR range. For example, if a chromium-forsterite femtosecond laser with a wavelength of $1.25 \,\mu\text{m}$ is used for pumping, then the wavelength of Stokes radiation will be 2.6 μm .

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