Quantum Electronics **46** (7) 581 – 585 (2016)

LASERS

DOI: 10.1070/QEL16007

1-kHz-repetition-rate femtosecond Raman laser

N.V. Didenko, A.V. Konyashchenko, P.V. Kostryukov, L.L. Losev, V.S. Pazyuk, S.Yu. Tenyakov

Abstract. **A femtosecond Raman laser utilising compressed hydrogen is experimentally investigated under pumping by radiation from a 1-kHz-repetition-rate Ti : sapphire laser. In the regime of double-pulse pumping, the conditions are determined, which correspond to the minimal energy dispersion of Stokes pulses. The optical scheme is realised, which is capable of ensuring the long-term stability of the average power of the first Stokes component with a variation of less than 2%. The Stokes pulses are produced with a pulse duration of 60 fs and energy of 0.26 mJ at a conversion efficiency of 14%.**

Keywords: *stimulated Raman scattering, femtosecond laser, pulse repetition rate, power stability.*

1. Introduction

Frequency conversion of radiation of femtosecond lasers based on stimulated Raman scattering (SRS) is an alternative to the employment of crystal parametric converters. In [1, 2], a method was developed for generating femtosecond pulses in SRS based on synchronisation of Raman components under biharmonic pumping. In these schemes, a series of low-energy femtosecond pulses separated by a ~10-fs time interval determined by the Stokes shift in an active medium is generated. For obtaining high-power single Stokes pulses, a scheme of SRS conversion of femtosecond pulses was proposed, according to which the pulse duration is initially increased by means of a frequency chirp and then SRS conversion of the chirped pulse is performed with the following compression [3]. This method allowed one to suppress concurrent nonlinear effects [4] such as self-focusing and self-phase modulation. Later this method has been improved by introducing double-pulse pumping, which eliminates narrowing of the Stokes spectrum as compared to the spectrum of the initial laser radiation pulse. Respectively, after compression, the Stokes pulse with a duration close to a laser pulse duration can be obtained [5].

In the scheme of chirped pulse conversion, femtosecond Raman lasers utilising compressed gases [6, 7] and crystals [4, 8, 9] have been studied. The shortest pulse duration of the first Stokes component was 40 fs in the case of SRS in compressed hydrogen of the radiation of a Ti : sapphire laser [7]. The maximal pulse energy of the first Stokes component (3 mJ) with a duration of 115 fs has been reached under SRS in a barium nitrate crystal [9] at a pulse repetition rate of 10 Hz.

In experimental investigations and some applied problems, Raman lasers can serve as frequency converters of radiation of widely used femtosecond laser systems. In particular, a Raman laser utilising compressed hydrogen can shift the radiation frequency of a Ti : sapphire laser to the wavelength range of 1.2 μm, where generation of THz radiation in organic crystals is quite efficient [10]. In the case of converting the radiation of a chromium-forsterite laser by SRS in hydrogen, a possibility arises to move to the mid-IR range (~2.5 μm) which is promising from the viewpoint of attosecond pulse generation [11]. In this case, sufficiently strong requirements are placed on SRS converters. In addition to the high conversion efficiency, they should provide a stable operation regime at an average output radiation power of ~1 W, a peak power of 1 – 10 GW and a pulse repetition rate of ~1 kHz. Presently, the maximal average output power of a femtosecond Raman laser reached in [9] is ~30 mW. However, the data on the operation stability of femtosecond Raman lasers are absent.

We present the results of an experimental study of a 1-kHz-repetition-rate femtosecond Raman laser utilising compressed hydrogen. The study is aimed at choosing the optical scheme and optimal parameters for a SRS converter, needed for obtaining the maximal operation stability at a high average power.

2. Experimental setup

The optical scheme of the experimental setup is presented in Fig. 1. A Ti : sapphire femtosecond laser system (REUS-40F1K) included a master oscillator and a regenerative amplifier and operated in the regime of chirped pulse amplification with the following time compression in a grating compressor. After compression, the pulse duration was 35 fs, the spectral width was 27 nm, the centre wavelength was 795 nm, the pulse energy was 2 mJ and the diameter of the beam with a Gaussian intensity distribution at the e^{-2} level was 8 mm. The pulse repetition rate could be varied from 100 to 1000 Hz. Since the pumping of a Raman laser needs chirped pulses of picosecond duration, the possibility of a smooth change in the output pulse duration was provided by varying the parameters of the grating compressor. For this purpose, the compressor grating was placed on a translation stage moved by a step motor. The movement of the grating by 0 – 1.5 cm pro-

N.V. Didenko, A.V. Konyashchenko, P.V. Kostryukov, L.L. Losev, V.S. Pazyuk P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia;
e-mail: dk_nick@mail.ru, lllosev@mail.ru, fs@avesta.ru;
S.Yu. Tenyakov Avesta Ltd, Fizicheskaya ul. 11, 142190 Troitsk, Moscow, Russia; e-mail: tenyakov@avesta.ru

Received 24 December 2015; revision received 31 May 2016
Kvantovaya Elektronika **46** (7) 581 – 585 (2016)
Translated by N.A. Raspopov

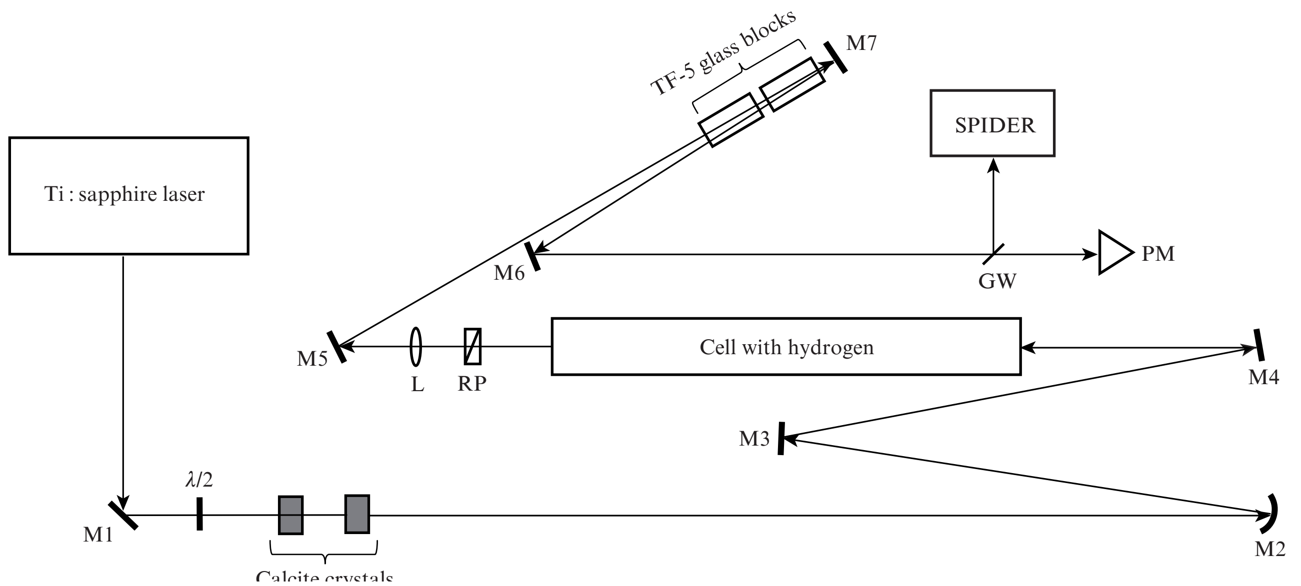

Figure 1. Optical scheme of experimental setup: (M1, M3, M4) plane mirrors (the reflection coefficient is 100% at the wavelength $\lambda = 800$ nm); (M2) mirror with a radius of curvature 5 m (100% at $\lambda = 800$ nm); (M5–M7) dichroic mirrors (100% at $\lambda = 1200$ nm and 20% at 800 nm); (GW) glass wedge; ($\lambda/2$) half-wave plate; (L) lens with a focal length of 1m; (RP) polarisation Rochon prism; (PM) power meter.

vided a smooth variation of the pulse duration from 35 fs to 5 ps.

In our previous works [5–8], two successive pulses with orthogonal linear polarisations needed for pumping a femtosecond Raman laser were formed by using the optical systems that comprised film polarisers and mirrors. Since the Raman laser is pumped by focused light beams with a divergence of $\sim 10^{-4}$ rad, the optical axes of the beams should be parallel with an accuracy of $\sim 10^{-5}$ rad in order to provide the spatial overlapping of the light beams in the focal waist region. At so high accuracy, small variations in the environment (temperature variations, mechanical perturbations and so on) may result in noticeable changes in the output parameters of the Raman femtosecond laser and reduce the long-term operation stability.

For increasing the stability we have chosen the scheme with birefringent crystals for forming two orthogonally polarised pulses. The time interval between the pulses was increased by placing two identical calcite crystals with a thickness of 1 cm and AR-coated faces in front of the cell with gaseous hydrogen. The principal optical axes of the crystals were parallel and lay in the planes parallel to the input faces. The crystals provided two orthogonally polarised pulses with a delay of about 10 ps between them. The ratio of pulse energies was varied by turning a half-wavelength plate placed in front of the crystals.

The laser radiation was focused by a mirror with a focal length of 2.5 m into the stainless steel cell filled with compressed hydrogen. The cell length was 1.2 m; its internal diameter was 1 cm.

Pulses with orthogonal polarisations at the output from the cell were separated by a polarisation Rochon prism. Then the light beam was collimated by a lens, was reflected from dichroic mirrors for selecting the radiation of first Stokes component and was passed to the time compressor.

The energy efficiency of the compressor was increased by choosing the scheme based on compression of a negatively chirped pulse propagating through a medium with normal dispersion. In our case, the dispersion medium was blocks of TF-5 glass. A double-pass scheme was used with a total medium length of 69 cm. In such a compressor with AR-coated faces of the optical blocks, a highly efficient compression can be obtained. Negligible losses are determined by scattering and absorption in the optical glass and by imperfect AR coatings.

The compressed pulse duration was measured with a SPIDER system (SP-120) modified to the radiation wavelength of 1.25 μm and based on the interferometric measurement of the electric field intensity. Statistical characteristics of the output radiation of the Raman laser were determined by using an Ophir Vega power meter with a pyroelectric sensor.

3. Discussion of experimental results

Experiments were performed at a 5-ps duration of a negatively chirped pulse at output from a laser system. For this purpose, an additional negative dispersion of -63000 fs^2 was introduced by shifting the grating in the output compressor by 13 mm relative to the position corresponding to the maximal pulse compression at the output from the regenerative amplifier. The choice of the pulse duration (5 ps) is explained by the necessity of a minimal laser pulse power for eliminating undesirable nonlinear effects (self-focusing, self-phase modulation), which suppress the SRS, and at the same time for providing the complete time separation of the pulses with orthogonal polarisations at a delay of 10 ps between them.

Issuing from the known nonlinear refractive index for hydrogen [12], the critical power of self-focusing is P_s[GW] = $12/p$ [atm], where p is the hydrogen pressure. Since the maximum pump power in the experiment did not exceed 0.4 GW (all the energy was concentrated in a single pulse with an energy of 2 mJ and duration of 5 ps), self-focusing had no effect on the SRS process at a hydrogen pressure of less than 30 atm.

For obtaining a high spatial quality of the light beam of the first Stockes component in the regime of transient SRS (in order to reproduce the wavefront of the pumping), the pump pulse energy should not exceed the threshold energy by a factor of above 1.5 [13]. Measurements of the energy dependence of the first Stokes component on the hydrogen pressure made it possible to find the pressure corresponding to the SRS threshold. At a pump pulse energy of 1.9 mJ and duration of 5 ps, the threshold hydrogen pressure was 12 atm. Since in transient SRS in hydrogen with a pressure of above 10 atm the threshold energy of SRS is inversely proportional to the pressure of hydrogen [14], at a pressure of 18 atm the pulse energy of 1.8 mJ will exceed the threshold energy by a factor of 1.5. Indeed, at such a hydrogen pressure, the Stokes light beam had a uniform intensity distribution over a transverse cross section. If the pressure increased to 25 atm, a small-scale modulation arose in the intensity distribution, and the conversion efficiency to the first Stockes component remained the same.

Figure 2 shows a dependence of the energy of the delayed pulse of the first Stockes component on the energy of the delayed pump pulse. One can see that in the range of 600 – 1800 μJ the Stokes pulse energy is actually constant and equals ~250 μJ, which corresponds to a conversion efficiency of 14%. In this range, the energy of the first pump pulse does not exceed 1200 μJ, which is below the threshold value, and all the energy of Stokes radiation is concentrated in the delayed pulse. At an energy of the delayed pulse below 600 μJ (the energy of the first pulse is 1200 μJ), the Stokes component arises in the first pulse as well. Note that the total energy of the first component is independent of the ratio of energies of the first and the delayed pump pulses because in a transient regime of SRS the process of generation is determined by the energy deposited into an active medium rather than by the pump radiation power (the pulse shape).

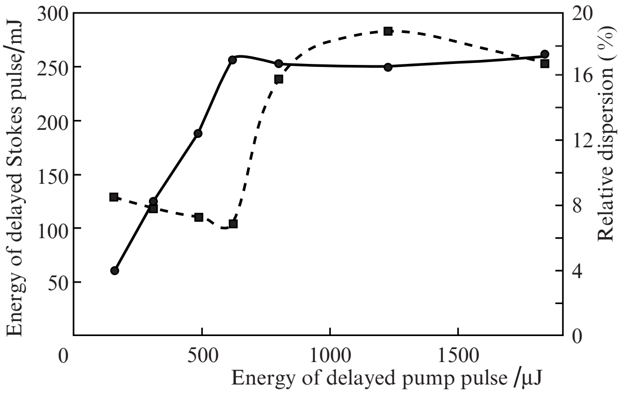

Figure 2. Energy of the delayed Stokes pulse (solid curve) and its relative dispersion (dashed line) vs. the energy of the delayed pump pulse.

Figure 2 also presents a relative dispersion (the ratio of the dispersion to the average value) of the energy of Stokes pulses versus the energy of the delayed pump pulse. The dispersion was measured in a series of 10^4 pulses. In the energy range, where the Stokes pulse arises only in the delayed pump pulse, the relative dispersion is 16% – 18%. The relative dispersion of the laser pulses is 0.5%. In transferring to the regime where the Stokes pulse also arises in the first pump pulse, the relative dispersion falls to 7% – 8%. Such a behaviour of the relative dispersion may be explained by the fact that if an intensive Stokes wave develops in the first pulse, and the delayed pump pulse scatters on an already formed phonon wave, then for the delayed pulse the processes related to quantum fluctuations of the initial stage of Raman scattering are eliminated [15, 16]. However, the relative energy dispersion of the first Stokes component is substantially higher than the dispersion of the pump pulses. In our opinion, the main contribution to the energy instability of the pulses of the first Stokes component is made by the generation of second-order Stokes radiation at a wavelength of 2.5 μm. A previously measured efficiency of this process, which limits the efficiency of conversion to the first Stokes component, reached 5% at the energy exceeding the first-order Stokes radiation threshold by a factor of 1.5 – 2 [17].

In the range of energies below 600 μJ, the energy efficiency of converting the delayed pump pulse into Stokes radiation reaches 42% (Fig. 2). At a greater energy, the Stokes component is generated by the first pump pulse. Correspondingly, the quantum efficiency approaches 63%. In the case of single-pulse excitation of the active medium the energy efficiency of conversion is only 14%. As a consequence, the spectrum of the Stokes pulse in the case of double-pulse pumping becomes wider (Fig. 3). As one can see, under pumping by single negatively chirped pulses, the high-frequency spectral components corresponding to the leading edge of a laser pulse are cut. Under double-pulse pumping, the spectrum of the Stokes component of the delayed pump pulse becomes symmetrical; its width reaches 44 nm (at half the maximum intensity), which corresponds to a transform-limited pulse with a duration of 48 fs.

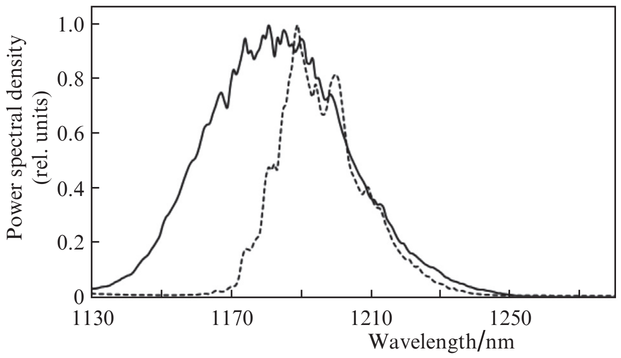

Figure 3. Spectrum of the Stokes pulse under single-pulse pumping (dashed curve) and of the delayed pulse under double-pulse pumping (solid curve).

Time compression of the Stokes pulse with a negative frequency chirp has been realised by passing radiation through TF-5 glass blocks. The double-pass scheme was used in this case. The total path length in glass was 69 cm. The second-order dispersion of the glass blocks was +62000 fs^2, which compensated for the negative dispersion introduced by the detuning of the grating compressor at the laser output. The dispersion characteristics of the pulse and the glass compressor were exactly matched by shifting the grating of the laser compressor. The pulse shape measured by the SPIDER system is shown in Fig. 4. The minimal pulse duration after compression was 60 fs at the 48-fs duration of a transform-limited pulse with a similar spectrum. Imperfect compression and arising post-pulses are mainly caused by the uncompensated third-order dispersion. This is confirmed by the dependence of the pulse phase on time (Fig. 4). The third- and higher-order dispersions can be compensated for by using an acousto-optical delay line [7].

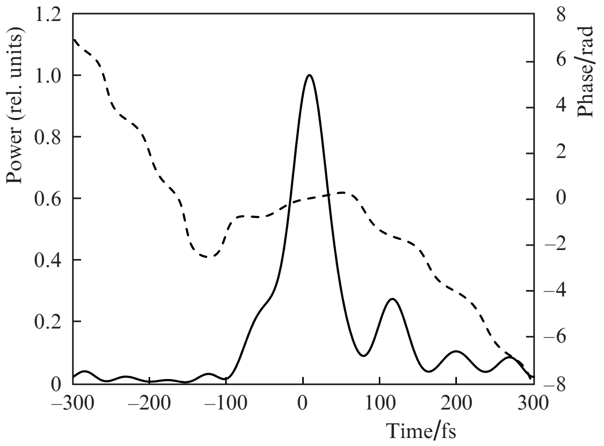

Figure 4. Profile (solid curve) and phase (dashed curve) of a Stokes pulse after compression.

Some applications of nonlinear converters of laser radiation require not only a small dispersion of the pulse energy, but also stability of their average energy (average radiation power under operation in the repetitively pulsed regime). This is necessary in performing experiments related to a large number of laser pulses (the regime of scanning or accumulation, laser micromachining of materials, and so on). At a high stability of the average power of the laser source, the power drift of a nonlinear converter is mainly caused by a change in external conditions, such as temperature, air flows and mechanical vibrations. As a result result, for example, the phase matching may worsen in parametric generators and harmonic generators, and the energy may be lost in introducing radiation into fibres and hollow waveguides.

The long-term stability is mainly enhanced by using sophisticated electro-mechanical feedback systems [18, 19]. As was already mentioned, the present work is aimed at minimising the influence of environment on the operation of a femtosecond Raman laser with double-pulse pumping. Of primary importance was to eliminate fluctuations of the angle between optical axes of the orthogonally polarised light beams. For this purpose, the optical system dividing the initial laser beam to the orthogonally polarised beams consisting of polarisers and mirrors [5–8] was substituted for birefringent crystals. Dependences of the average radiation power of a Ti:sapphire laser and of the first Stokes component on time are shown in Fig. 5. In measurements, the power was averaged over 65 pulses. One can see that the variation in the average laser power over the total measurement range (variation swing) was ~10 mW at an average power of ~1.9 W. The corresponding relative variation in the average power was ~0.5%. In the Raman laser this variation was greater, but did not exceed 2.5% (5 mW at an average power of 260 mW). As compared to our previous investigations [5–8] of femtosecond Raman lasers, the relative variation in the average power is lower by more than an order of magnitude.

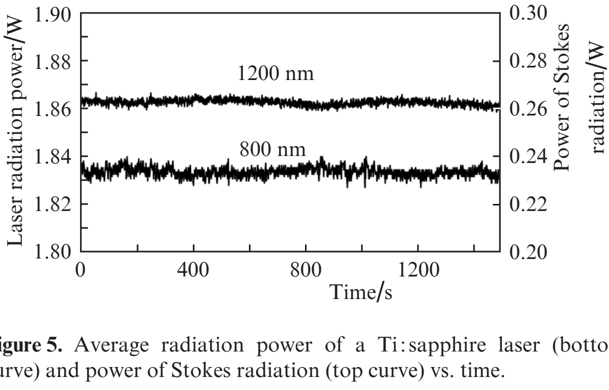

Figure 5. Average radiation power of a Ti:sapphire laser (bottom curve) and power of Stokes radiation (top curve) vs. time.

4. Conclusions

Thus, investigations of a 1-kHz-repetition-rate femtosecond Raman laser utilising compressed hydrogen have made it possible to find the regime in which the minimal relative energy dispersion of pulses of the first Stokes component is 7%. The pulses of Stokes radiation with a duration of 60 fs and energy of 0.26 mJ have been obtained at an energy conversion efficiency of 14%. The average power of output radiation is 0.26 W.

The scheme of double-pulse excitation of the active medium based on splitting the initial laser beam into two orthogonally polarised beams by birefringent crystals, which provides automatic matching of the light beams, has allowed us to weaken the influence of environment factors on the operation stability of the converter and to reach the relative variation in the average power of the first Stokes component at a level of ~2%.

Note that in reducing the pulse repetition rate to 100 Hz with other parameters being the same, the conversion efficiency increases to 20%, which points to an influence of gas heating on the SRS generation. This circumstance opens possibilities for increasing the conversion efficiency and reducing the duration of a Stokes radiation pulse while operating at a pulse repetition rate of 1 kHz by using schemes with a gas exchange in the conversion zone.

References

1. Zaitsu S., Kida Y., Imasaka T. *Phys. Rev. A*, **70**, 031801 (2004).
2. Sali E., Mendham K., Tisch J., Halfman T., Marangos J.P. *Opt. Lett.*, **29**, 495 (2004).
3. Zhavoronkov N., Noack F., Petrov V., Kalosha V.P., Herrmann J. *Opt. Lett.*, **26**, 47 (2001).
4. Bespalov V.G., Efimov Y.N., Staselko D.I., Krylov V.N., Ollikainen O., Wild U.P., Rebane A. *Proc. SPIE Int. Soc. Opt. Eng.*, **4069**, 154 (2000).
5. Konyashchenko A.V., Losev L.L., Tenyakov S.Yu. *Opt. Express*, **15**, 11855 (2007).
6. Konyashchenko A.V., Losev L.L., Pazyuk V.S., Tenyakov S.Yu. *Appl. Phys. B*, **93**, 455 (2008).
7. Didenko N.V., Konyashchenko A.V., Kostryukov P.V., Losev L.L., Pazyuk V.S., Tenyakov S.Yu., Molchanov S.Ya., Chizhikov S.I., Yushkov K.B. *Kvantovaya Elektron.*, **45**, 1101 (2015) [*Quantum Electron.*, **45**, 1101 (2015)].
8. Konyashchenko A.V., Losev L.L., Tenyakov S.Yu. *Kvantovaya Elektron.*, **40**, 700 (2010) [*Quantum Electron.*, **40**, 700 (2010)].
9. Grigsby F.B., Peng Dong, Downer M.C. *J. Opt. Soc. Am. B*, **25**, 346 (2008).
10. Vicario C., Jazbinsek M., Ovchinnikov A.V., Chefonov O.V., Ashitkov S.I., Agranat M.B., Hauri C.P. *Opt. Express*, **23**, 4573 (2015).
11. Chen M.-C., Arpin P., Popmintchev T., Gerrity M., Zhang B., Seaberg M., Popmintchev D., Murnane M.M., Kapteyn H.C. *Phys. Rev. Lett.*, **105**, 173901 (2010).
12. Lehmeier H.J., Leupacher W., Penzkofer A. *Opt.Commun.*, **56**, 67 (1985).

13. Zel'dovich B.Ya., Pilipetskii N.F., Shkunov V.V. *Principles of Phase Conjugation* (Berlin: Springer, 1985; Moscow: Nauka, 1985).
14. Shen Y.R. *The Principles of Nonlinear Optics* (New York: Wiley, 1984; Moscow: Nauka, 1989).
15. Raymer M.G., Mostowski Y. *Phys. Rev. A*, **24**, 1980 (1981).
16. Apanasevich P.A., Gakhovich D.E., Grabchikov A.S., Kilin S.Ya., Kozich V.P., Kontsevoi B.L., Orlovich V.A. *Kvantovaya Elektron.*, **19**, 884 (1992) [*Quantum Electron.*, **22**, 822 (1992)].
17. Konyashchenko A.V., Losev L.L., Tenyakov S.Yu. *Kvantovaya Elektron.*, **41**, 459 (2011) [*Quantum Electron.*, **41**, 459 (2011)].
18. Seres J., Muller A., Seres J., O'Keeffe K., Lenner M., Herzog R.F., Kaplan D., Spielmann C., Krausz F. *Opt. Lett.*, **28**, 1832 (2003).
19. Ricci A., Jullien A., Rousseau J., Lopez-Martens R. *Appl. Sci.*, **3**, 314 (2013).