Quantum Electronics **52** (3) 278–282 (2022)

https://doi.org/10.1070/QEL18002

Efficient SRS of chirped Ti : sapphire laser pulses in $BaWO_4$ crystals

I.O. Kinyaevskiy, V.I. Kovalev, A.V. Koribut, Ya.V. Grudtsyn,
L.V. Seleznev, E.E. Dunaeva, A.A. Ionin

Abstract. **We report an experimental study of SRS of chirped 40-ps Ti : sapphire laser pulses with a transform-limited duration of 0.35 ps and centre wavelength of 925 nm in $BaWO_4$ crystals. The highest generation efficiency for the ~925 cm^{-1} shifted Stokes signal of SRS by the v_1 vibrational mode of $BaWO_4$ crystals has been reached using three sequentially located crystals with a total length of 3.3 cm. The position of the crystals after a focusing mirror has been optimised, which has ensured a pump-to-Stokes conversion efficiency of ~50% in terms of spectral brightness and ~20% in terms of pulse energy.**

Keywords: *stimulated Raman scattering, $BaWO_4$ crystal, Ti : sapphire laser, femtosecond pulse.*

1. Introduction

Stimulated Raman scattering (SRS), a nonlinear optical effect, was discovered in 1962 [1], i.e. about a year after the discovery of second harmonic generation [2]. At present, this effect is unrivalled in its range and diversity of applications for resolving scientific and applied issues. They include a considerable extension of the capabilities of conventional Raman spectroscopy [3, 4]; laser light conversion with the aim of expanding into new spectral ranges and controlling the pulse duration and shape, as well as improving pulse energetics and directionality [5–10]; cooling of atoms [11]; wave front conjugation [12]; and a large number of others.

In the late 1960s, with the advent of lasers capable of generating pulses with a duration comparable to (or shorter than) the relaxation time of Raman-active vibrations involved in SRS interaction, a new research direction emerged, known as transient SRS. Its theoretical basis was provided by Khokhlov and his colleagues (see e.g. Ref. [13]) and Bloembergen et al. [14]. Despite the more than half-a-century history of research and development in this area of nonlinear optics, interest in it and its topicality persist and are maintained by both the nontrivial physics of the processes involved in such interaction and interesting applications. In particular, a Raman converter is currently regarded as a key component of promising all-solid-state mid- and far-IR subpico- and femtosecond laser pulse sources intended, e.g. for injection of their output into high-power CO_2 laser amplifiers [15–17].

In addressing this issue, $BaWO_4$ crystals were examined as an active medium of Raman converters [16, 17]. The choice of this material was prompted by the facts that, first, the frequency of its strongest Raman-active phonon mode, with a wavenumber $v_1 \approx 925$ cm^{-1} [18], lies in the gain band and is close to the peak of the gain profile of high-pressure CO_2 laser amplifiers ($v \approx 940$ cm^{-1} and wavelength $\lambda \approx 10.6$ μm) and that, second, it has an almost record large SRS gain coefficient ($g \approx 40$ cm GW^{-1} at a pump wavelength $\lambda_p \approx 0.53$ μm [19]). The decay time of the v_1 mode, $T_2 = 6.6$ ps [18], considerably exceeds the duration of the pulses of interest for us, so SRS interaction in it is essentially transient. In such a case, the pump intensity needed for noticeable stimulated Raman conversion is considerably higher, and an important role is played by other nonlinear effects, which often have a negative impact on the stimulated Raman conversion process. Because of this, in most known studies dealing with SRS of ultrashort laser pulses in SRS-active media the highest conversion efficiency achieved did not exceed a few percent. At the same time, in $BaWO_4$ crystals we obtained an unusually high efficiency of the stimulated Raman conversion of visible femtosecond pump pulses with $\lambda_p = 0.47–0.515$ μm to the first Stokes [16, 17]. In particular, the efficiency of the stimulated Raman conversion of frequency-doubled ytterbium fibre laser pulses (pump pulse duration $t_p = 300$ fs, $\lambda_p = 515$ nm) to the first Stokes, shifted by ~925 cm^{-1} relative to the pump frequency, reached ~20% [16]. It was planned to convert the two-colour pulses thus obtained [16, 17] to the 10-μm range by difference frequency generation (DFG), a variation of the optical rectification method proposed by Akhmanov and Khokhlov [20]. However, attempts to perform such conversion in $LiGaS_2$ nonlinear crystals, essentially the only available material transparent both at $\lambda_p \leqslant 0.45$ μm and in the 10-μm range [21], were unsuccessful, primarily because the crystals had an insufficient second-order nonlinear susceptibility and optical damage threshold. In view of this, to ensure DFG it is reasonable to use longer wavelength two-colour light, which considerably increases the number of potentially applicable nonlinear crystals, including those with a higher nonlinear susceptibility and optical damage threshold [22].

Unfortunately, producing longer wavelength two-colour light via SRS is complicated by the fact that, with increasing pump wavelength λ_p at constant T_2, the SRS gain coefficient decreases in proportion to λ_p^2 [23] and, hence, the pump intensity needed to achieve sufficiently high stimulated Raman conversion efficiency increases. Accordingly, this increases the role of concomitant nonlinear processes, such as self-

I.O. Kinyaevskiy, V.I. Kovalev, A.V. Koribut, Ya.V. Grudtsyn, L.V. Seleznev, A.A. Ionin Lebedev Physical Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: kinyaevskiyio@lebedev.ru;
E.E. Dunaeva Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia

Received 15 November 2021
Kvantovaya Elektronika **52** (3) 278–282 (2022)
Translated by O.M. Tsarev

phase modulation, avalanche ionisation, and optical breakdown, which limit the attainable stimulated Raman conversion efficiency [24, 25]. The problem can be obviated by using chirped pump pulses for SRS excitation [26–28]. To this end, the input ultrashort chirped laser pulse duration should be increased using dispersing elements, and an SRS-converted, stretched pulse can then be recompressed to its transform-limited duration. In this work, we examine the feasibility of reaching high efficiency of stimulated Raman conversion to the first Stokes with $\nu_1 \approx 925$ cm^{-1} in a group of $BaWO_4$ crystals under pumping with chirped Ti:sapphire laser pulses stretched to 40 ps.

2. Experimental configuration

Our experiments were carried out at the Center for Laser and Nonlinear Optical Technologies, N.G. Basov Quantum Radiophysics Division, Lebedev Physical Institute, Russian Academy of Sciences (RAS), with the use of the Start-480M Ti:sapphire laser system (Avesta Project Ltd., Russia). The optical layout of the experiments is shown in Fig. 1. The system, comprised of a master oscillator, stretcher, regenerative amplifier, and compressor, was adjusted so as to generate 925-nm pulses with a spectral width at half maximum of 3.5 ± 0.5 nm, energy of up to 11 mJ, and duration of 0.35 ± 0.05 ps after the optical compressor. The 40-ps chirped laser pulses used in this study were extracted from the system before compression in the optical compressor.

The amplified chirped laser pulses of ~40 ps duration were focused by an $f = 1$ m spherical mirror (represented as a lens in Fig. 1). The $1/e^2$ laser beam diameter on the mirror was 0.9 cm. The Raman converter consisted of three $BaWO_4$ crystals mounted sequentially after the mirror. This configuration made it possible to compensate for the decrease in pump intensity I_p as a result of the linear optical loss in the crystals. Samples 1 (1.5 cm long) and 2 (1 cm long) were located 60 and 80 cm from the mirror, respectively, and sample 3 (0.8 cm long) was translated along the convergent pump beam, which allowed us to vary the intensity inside it. The distances between the samples and mirror were chosen so that their surfaces remained intact even at the highest pump energy. The longest samples were placed first in order to ensure the highest SRS gain for the Stokes signal during its exponential growth.

The incident light was polarised across the optical c-axis of the crystals. After passing through the samples, the light was attenuated by reflecting it near normal incidence from two glass wedges and directed to an ASP-150 spectrometer (Avesta Project Ltd., Russia). The noise level of the detection system allowed us to identify the sought Stokes signal with an amplitude at a level of at least 0.2% of the pump amplitude. The $BaWO_4$ crystals were grown and prepared for this study at the Prokhorov General Physics Institute, RAS.

3. Experimental results

Without sample 3 in the configuration studied, no Stokes signal was detected near 1010 nm even at the highest energy of incident pulses, 11 mJ. If sample 3 was placed 85 cm from the spherical mirror, at a pulse energy of 8.0 ± 0.5 mJ such a signal was detected and its amplitude was ~0.8% of the pump amplitude (Fig. 2a). Increasing the pump pulse energy to 11.0 ± 0.5 mJ increased the peak amplitude by more than one order of magnitude (Fig. 2b). The internal Stokes signal generation efficiency was then 12% in terms of amplitude (spectral brightness) and 2.3% in terms of energy.

In a generally accepted model for SRS from a spontaneous scattering level [29], the gain increment $G = gI_p l$ that ensures ~1% conversion efficiency (threshold increment G_{th}) in steady-state interaction mode is ~25. In our conditions, the total SRS gain increment G for all three samples was calculated as

$$G = g_0(I_1 l_1 + I_2 l_2 + I_3 l_3), \tag{1}$$

where $g_0 = 10$ cm GW^{-1} is the steady-state SRS gain coefficient at $\lambda_p \cong 925$ nm [18] and $I_{1,2,3}$ is the intensity incident on the samples of length $l_{1,2,3}$.

The total threshold SRS gain increment G_{th} for the three $BaWO_4$ crystals, evaluated from the present experimental data on the pulse energy and duration, the incident beam

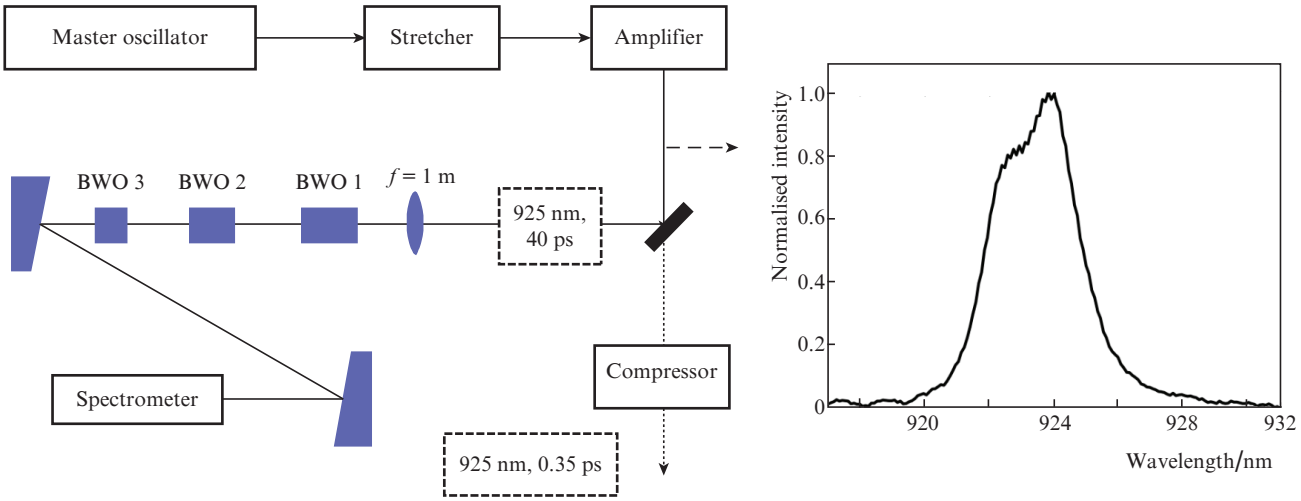

Figure 1. Optical layout of the experiments. The right panel shows the spectrum of an amplified laser pulse.

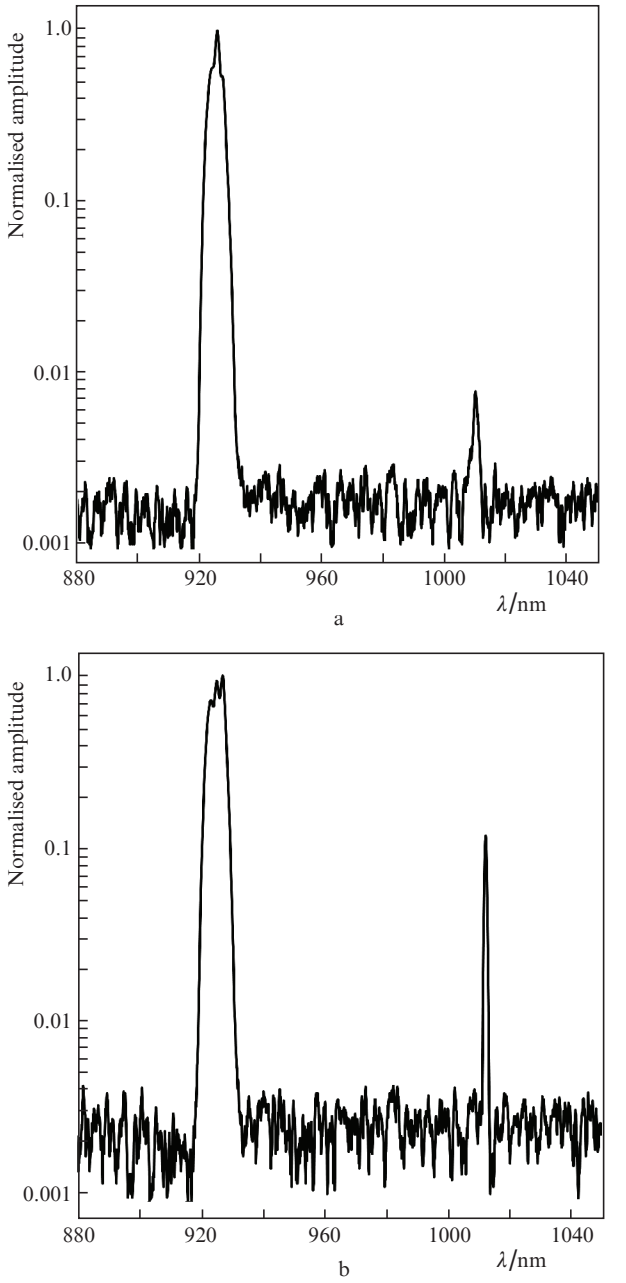

Figure 2. Spectra of pulses passed through three $BaWO_4$ crystals at a pump pulse energy E_p = (a) 8 and (b) 11 mJ.

diameter, the length of the samples, and the optical loss in them, was found to be far above 25: ~300 at E_p = 8 mJ and ~400 at 11 mJ (in the case of two samples, the gain increment at the highest pulse energy was ~200). A similar increase in threshold SRS gain increment for chirped pulses was observed previously as well [26–28]. The cause of this considerable increase in threshold increment is that, even though SRS interaction in our case can be thought of as an almost steady-state process (the pump pulse duration, 40 ps, is almost one order of magnitude longer than time T_2), the frequency of the pump emission spectrum varies with time during a pulse.

The fact that pump pulse chirping reduces the SRS gain coefficient is a consequence of the finite decay time of molecular vibrations, i.e. of the nonzero time T_2. The point is that, during SRS, the force producing vibrations originates from beating of the pump field (at a frequency ω_p) and scattered field (at a frequency ω_{sc}), the latter originating from the former [23]. The variation of the pump frequency with time leads to a variation in the instantaneous frequency of the force producing vibrations. This means that, if at some instant in time the frequency of this force, $\Omega = \omega_p - \omega_{sc}$, coincides with the resonance frequency of the oscillator, Ω_r, it turns out to be detuned from the resonance in the next instant. Clearly, the vibration build-up efficiency decreases in such a case [30]. Since in the case of SRS it is the amplitude of the vibrations being built-up which determines the SRS gain coefficient g_r [23], the dependence of the amplitude of such vibrations on frequency detuning, $\Delta\Omega = \Omega_r - \Omega$, is just the expected effect. Analysis of the response of the medium in the case of SRS as a forced oscillation in a weakly nonlinear dissipative system [30] yields g_r as a function of frequency detuning for chirped pulses [31]:

$$g_{r,ch} = g_r/[1 + (\Delta\Omega T_2/2)^2], \quad (2)$$

where in our case $\Delta\Omega = CT_2$ is the frequency detuning of chirped pulses over time T_2, and C is the chirp parameter. At $\Delta\lambda_p$ = 3 nm, t_p = 40 ps, and $|C| \cong 1.6 \times 10^{23}$ s^{-2}, it follows from (2) that $g_{r,ch} \cong g_r/12$. The increase in threshold increment observed in our experiments agrees well with this estimate. The fact that increasing E_p from 8 (SRS threshold in our case, see above) to 11 mJ increased the conversion efficiency by 15 times, rather than by four orders of magnitude, as would be expected in the case of exponential growth, is a consequence of SRS gain saturation [29].

Note also that, along with the increase in threshold increment due to pump pulse chirping, the peak of the Stokes SRS spectrum was observed to shift with increasing E_p. In particular, increasing E_p from 8 to 11 mJ changed the spectral position of the Stokes peak from 1010 to 1013 nm, which corresponds to Raman shifts of 900 and 925 cm^{-1}, respectively. A similar effect in the case of the SRS of positively chirped ($C > 0$) laser pulses was observed in $Ba(NO_3)_2$ crystals as well [27]. Basic to the observed effect is the fact that, near the SRS threshold, Stokes generation begins with a long delay relative to the leading edge of the pump pulse and decays essentially simultaneously with the pump pulse [14]. With increasing gain increment, the rate of Stokes pulse development increases, leading to a decrease in the delay of the leading pulse edge relative to the pump pulse edge [14]. As a result, in the case of chirped pump pulses the observed frequency of the scattered signal is determined primarily by the frequency of the pulse during its decay. As the pumping level increases, the delay decreases and, at a sufficiently long pump pulse duration, the scattered light pulse essentially reproduces the pump pulse shape and, accordingly, the pump spectrum.

Such a mechanism of the variation in Stokes frequency with pump energy is well consistent with results reported by Chizhov et al. [32], who observed a shift in response to changes in chirped pump pulse duration. They also showed that the Stokes shift was below the "resonance" frequency of the SRS-active mode (OH band, $\nu \cong 3400$ cm^{-1} in water [32]) at a negative chirp ($C < 0$) and above it at a positive chirp ($C > 0$). The fact that, in our experiments, the frequency shift decreased with decreasing E_p indicates, among other things, that in this study, like in a previous one [27], the chirp was positive.

Our attempts to raise the SRS coefficient by raising the pump intensity and, accordingly, the SRS gain increment G in sample 3 by shifting it closer to the focal waist of the convergent laser beam ensured no marked effect and resulted in optical damage of its backside at a light intensity below the expected threshold. One possible reason for this is the accelerated self-focusing of the convergent beam. In view of this, we made a series of measurements in which sample 3 was placed behind the focal plane. Translating the sample along the beam and thus varying the light intensity in it, we found its position corresponding to the highest Stokes SRS power (Fig. 3).

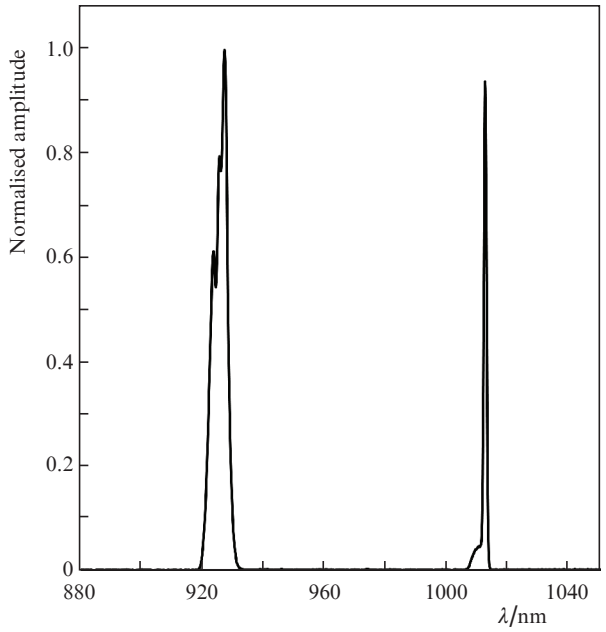

Figure 3. Spectrum of a laser pulse passed through the $BaWO_4$ crystals in the case when sample 3 is located behind the focal plane of the beam and the gain increment is $G \approx 300$.

The optimal separation between sample 3 and the focusing mirror was found to be 120 cm. This resulted in an integral SRS gain increment G calculated using (1) of about 300, and the Stokes signal and pump pulse had roughly equal spectral brightnesses at the output of sample 3, which corresponded to an internal conversion efficiency of ~50% (with no allowance for Fresnel reflection from the three crystals). It follows from Fig. 3 that the spectral width of a transmitted pump pulse increased from about 3.5 to 5 nm, the peak wavelength of the Stokes signal was near 1013 nm (frequency shift of ~925 cm^{-1}), and its spectral width was a factor of 4 smaller than the width of the pump spectrum. As a consequence, the internal stimulated Raman conversion efficiency in terms of energy is ~20%, and the potentially attainable transform-limited pulse duration is ~1 ps.

Thus, it has been shown that placing sample 3 behind the focal plane of the laser beam allows one to obtain a higher Stokes generation efficiency than in the case when all SRS crystals are located before the focal plane. We suppose that this is related to the interaction geometry and the effect of self-focusing on it. In the case of a convergent pump beam, the intensity at the crystal backside is higher than that at the frontside. Moreover, self-focusing further increases the difference, leading to optical damage of the backside. In the case of a divergent beam, self-focusing partially compensates for the beam divergence due to focusing the mirror, leading to a more uniform intensity distribution along the length of the crystal and, hence, a more efficient use of the entire sample length.

4. Conclusions

We have experimentally demonstrated efficient conversion of chirped 40-ps Ti:sapphire laser pulses to a Stokes signal at ~925 cm^{-1} via SRS in three $BaWO_4$ crystals with a total length of 3.3 cm. The crystals were located after a mirror that focused the pump beam. The highest conversion efficiency was observed when two crystals were located in front of the focal plane of the mirror, and one crystal was placed behind it. The Stokes signal and laser pulse had equal spectral brightnesses at the output of the crystals, which corresponded to an internal conversion efficiency of ~50%. The spectral width of the Stokes pulse was a factor of 4 smaller than that of the pump laser pulse, which corresponded to an internal stimulated Raman conversion efficiency of ~20% in terms of energy. In our experiments, the Stokes signal was observed to shift from 1010 to 1013 nm with increasing exponential SRS gain coefficient. The two-colour light obtained in this study can be used for ultrashort mid-IR pulse generation via difference frequency generation in second-order nonlinear crystals.

References

1. Woodbury E.J., Ng W.K. *Proc. IRE*, **50**, 2347 (1962).
2. Franken P.A., Hill A.E., Peters C.W., Weinreich G. *Phys. Rev. Lett.*, **7**, 118 (1961).
3. Akhmanov S.A., Koroteev N.I. *Metody nelineinoi optiki v spektroskopii rasseyaniya sveta* (Nonlinear Optical Methods in Light Scattering Spectroscopy) (Moscow: Nauka, 1981).
4. Akhmanov S.A. *Hyperfine Interact.*, **38**, 553 (1987).
5. Grasyuk A.Z. *Sov. J. Quantum Electron.*, **4**, 269 (1974) [*Kvantovaya Elektron.*, **1**, 485 (1974)]; *Tr. Fiz. Inst. im. P.N. Lebedeva, Akad. Nauk SSSR*, **76**, 75 (1974).
6. Grasiuk A.Z., Zubarev I.G. *Appl. Phys.*, **17**, 211 (1978).
7. Losev L.L., Soskov V.I. *Opt. Commun.*, **135**, 71 (1997).
8. Dianov E.M., Prokhorov A.M. *IEEE J. Sel. Top. Quantum Electron.*, **6**, 1022 (2000).
9. Bufetov I.A. Doct. Diss. (Moscow, General Physics Inst., 2002).
10. Zheltikov A.M. *Usp. Fiz. Nauk*, **181**, 33 (2011).
11. Reichel J., Bardou F., Dahan M.B., Peik E., Rand S., Salomon C., Cohen-Tannoudji C. *Phys. Rev. Lett.*, **75**, 4575 (1995).
12. Zel'dovich B.Ya., Mel'nikov N.A., Plipetskii N.F., Ragul'skii V.V. *Pis'ma Zh. Eksp. Teor. Fiz.*, **25**, 41 (1977).
13. Akhmanov S.A., Drabovich K.N., Sukhorukov A.P., Chirkin A.S. *Sov. Phys. JETP*, **32** (2), 266 (1971) [*Zh. Eksp. Teor. Fiz.*, **59**, 485 (1970)].
14. Carman R.L., Shimizu F., Wang C.S., Bloembergen N. *Phys. Rev. A*, **2**, 60 (1970).
15. Welch E.C., Tochitsky S.Y., Pigeon J.J., Joshi C. *Opt. Express*, **26**, 5154 (2018).
16. Kinyaevskiy I.O., Kovalev V.I., Danilov P.A., Smirnov N.A., Kudryashov S.I., Seleznev L.V., Dunaeva E.E., Ionin A.A. *Opt. Lett.*, **45**, 2160 (2020).
17. Kinyaevskiy I.O., Koribut A.V., Grudtsyn Y.V., Dunaeva E.E., Andreev Y.M., Ionin A.A. *Laser Phys. Lett.*, **17**, 105402 (2020).
18. Cerny P., Jelınkova H., Zverev P.G., Basiev T.T. *Prog. Quantum Electron.*, **28**, 113 (2004).
19. Lisinetskii V.A., Rozhok S.V., Bus'ko D.N., Chulkov R.V., Grabtchikov A.S., Orlovich V.A., Zverev P.G. *Laser Phys. Lett.*, **2**, 396 (2005).

20. Akhmanov S.A., Khokhlov R.V. *Problems of Nonlinear Optics* (Electromagnetic Waves in Nonlinear Dispersive Media) (New York: Gordon and Breach, 1972; Moscow: VINITI, 1964).
21. Isaenko L., Yelisseyev A., Lobanov S., Krinitsin P., Petrov V., Zondy J.-J. *J. Non-Cryst. Solids*, **352**, 2439 (2006).
22. Nikogosyan D.N. *Sov. J. Quantum Electron*., **7**, 1 (1977) [*Kvantovaya Elektron*., **4**, 5 (1977)].
23. Shen Y.R. *The Principles of Nonlinear Optics* (New York: Wiley, 1984; Moscow: Nauka, 1989).
24. Grabtchikov A.S., Chulkov R.V., Orlovich V.A., Schmitt M., Maksimenko R., Kiefer W. *Opt. Lett*., **28**, 926 (2003).
25. Konyashchenko A.V., Losev L.L., Pazyuk V.S. *Quantum Electron*., **51**, 217 (2021) [*Kvantovaya Elektron*., **51**, 217 (2021)].
26. Jordan C., Stankov K.A., Marowsky G., Canto-Said E.J. *Appl. Phys. B*, **59**, 471 (1994).
27. Zhavoronkov N., Noack F., Petrov V., Kalosha V.P., Herrmann J. *Opt. Lett*., **26**, 47 (2001).
28. Kinyaevskiy I.O., Seleznev L.V., Koribut A.V., Dunaeva E.E., Andreev Yu.M., Ionin A.A. *Izv. Vyssh. Uchebn. Zaved., Fiz*., **64**, 67 (2021).
29. Shen Y.R., Bloembergen N. *Phys. Rev*., **137**, A1787 (1965).
30. Migulin V., Medvedev V., Mustel E., Parygin V. *Basic Theory of Oscillations* (Moscow: Mir, 1983; Moscow: Nauka, 1978).
31. Boyd R.W. *Nonlinear Optics* (NY: Academic Press, 2007).
32. Chizhov P.A., Grishin M.Ya., Pershin S.M., Lednev V.N., Ushakov A.A., Bukin V.V. *Opt. Lett*., **46**, 2686 (2021).