Optics and Spectroscopy, 2023, Vol. 131, No. 2

06

Influence of focusing on transient SRS self-seed by SPM of 0.3 ps laser pulses in a $BaWO_4$ crystal

© I.O. Kinyaevskiy[1], A.V. Koribut[1,¶], I.V. Gritsenko[1], A.M. Sagitova[1], M.V. Ionin[1], E.E. Dunaeva[2], A.A. Ionin[1]

[1] Lebedev Physical Institute, Russian Academy of Sciences, 119991 Moscow, Russia
[2] Prokhorov Institute of General Physics, Russian Academy of Sciences (IOFAN), 119991 Moscow, Russia
[¶]e-mail: andrew-koribut@yandex.ru

Received December 20, 2022
Revised January 13, 2023
Accepted January 28, 2023

The effect of focusing, taking into account self-focusing, on the interference of SRS (stimulated Raman scattering) and self-phase modulation in a 8 mm $BaWO_4$ crystal pumped by laser pulses with a duration of 0.3 ps and a wavelength of 515 nm is experimentally studied. The maximum efficiency of SRS conversion ($\sim 23\%$) to the Stokes component of the $\nu_1 = 925\,\mathrm{cm}^{-1}$ strongest mode is obtained with a lens with a focal length of 40 mm at the linear focus shift towards the rear facet of the crystal. The increase in efficiency, when the linear focus is shifted to the rear facet, is associated with an increase in the distance between the linear and nonlinear foci, which results in an increase in the effective length of the nonlinear interaction.

Keywords:: stimulated Raman scattering, self-phase modulation, self-focusing, $BaWO_4$, femtosecond pulses.

DOI: 10.61011/EOS.2023.02.55784.6-23

Introduction

Laser emission conversion by nonlinear optics methods is widely used for obtaining the required (for a specific task) spectral radiation characteristics that are unavailable directly from laser sources. Multiple nonlinear optics methods are used: parametric conversion in quadratic nonlinear crystals [1], stimulated Raman scattering (SRS) [2], self-phase modulation (SPM) [3] and other. Constructive interference of SPM and SRS effects was found earlier in $BaWO_4$ (BWO) crystal during pumping with 0.3 ps 515 nm [4,5] laser pulses. Such interaction of two nonlinear effects allowed to achieve SRS stokes component generation efficiency which is unusually high for subpicosecond laser pulses in a simple single-pass optical configuration: up to $\sim 20\%$ on mode $\nu_1 = 925\,\mathrm{cm}^{-1}$ [4] and up to $\sim 35\%$ on mode $\nu_2 = 330\,\mathrm{cm}^{-1}$ [5]. Unusually high efficiency was achieved due to the fact that SPM effect formed seed emission for SRS offset to the appropriate Raman scattering mode frequency of BWO crystal and SRS effect enhanced these components.

SRS studies [4,5] and the relevant investigations of Kerr nonlinearity response of BWO crystal [6,7] were performed in the same optical configuration with 35–40 mm lens focusing. However, the interaction geometry influence defined both by the focusing numerical aperture and linear focus position inside the crystal focus position has not been investigated before. Therefore the purpose of the study was to determine the SRS conversion efficiency with SPM self-seed in BWO crystal with laser beam focusing parameter variation.

Experimental setup

Experiments for the investigation of SRS conversion efficiency of femtosecond laser pulses in BWO crystal were performed in the Center of Laser and Nonlinear Optical Technology (LPI). A 8 mm long crystal sample. was grown by Czochralski method from melt in air in the Institute of General Physics of the Russian Academy of Sciences (Department of Laser Materials and Photonics). The optical scheme of the experiment is shown in Figure 1. For crystal pumping, Satsuma (Amplitude Systemes) ytterbium ion based second harmonics optical fiber laser emission was used. Laser emission parameters are as follows: central wavelength 515 nm, pulse width 300 fs, pulse energy up to $3.1\,\mu$J, pulse repetition rate 1 kHz, beam diameter 1.7 mm (at level $1/e^2$). To vary the focusing numerical aperture, 40 mm and 90 mm lens and 20 mm lens system were used. It should be noted that the lens system introduced additional reflection loss (20%) compared with the lens, therefore the maximum energy of the laser pulse incident on the crystal in the experiments using the lens system was not higher than $2.5\,\mu$J. A 90 mm lens was places behind the crystal to transfer the image of the rear crystal face onto ASP-150 FT spectrometer entrance slit (spectral resolution 0.3 nm, Avesta Project Ltd). Optical c-axis of the crystal was perpendicular to the radiation polarization. To vary the linear focus position inside the crystal, the latter was secured

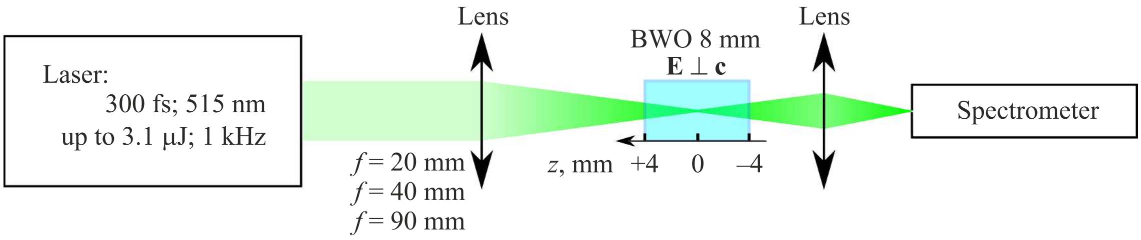

Figure 1. Optical scheme of the experiment. The z axis points out the linear focus position inside the crystal.

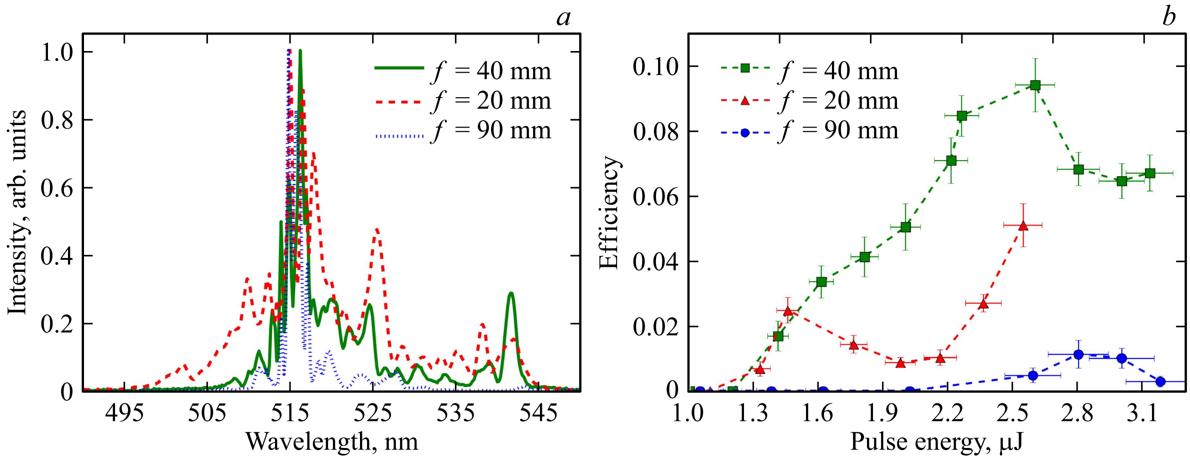

Figure 2. (a) Spectra of the emission passing through BWO crystal with maximum SRS component generation efficiency $925\,\text{cm}^{-1}$ at $z = 0$ mm and various focusing lengths: $f = 20$, 40 and 90 mm. (b) SRS component generation efficiency $\nu_1 = 925\,\text{cm}^{-1}$ of BWO crystal depending on the pulse energy at $f = 20$, 40 and 90 mm.

on a moving platform driven by a stepper motor. Linear focus position inside the crystal is determined on the z axis and measured in millimeters (Figure 1); $z = 0$ corresponds to the focus position in the crystal center, negative z values correspond to the linear focus displacement towards the rear face.

Focusing length influence

Figure 2 shows the examples of emission spectra and SRS generation efficiency of the strongest SRS mode of BWO crystal ($\nu_1 = 925\,\text{cm}^{-1}$) depending on the laser pulse energy for various focusing length at $z = 0$ mm. Conversion efficiency was defined in the same way as in [4]: as SRS component outline area vs. full spectrum area of the passing laser pulse. Energy measurement errors correspond to the laser system stability and are calculated as rms deviation of laser pulse energy at the laser system output. The efficiency measurement error is associated with the measurement accuracy of amplitudes and SRS component position boundaries. It should be noted that the ratios of Rayleigh length ($2\pi n\omega_o^2/\lambda$, where ω_0 is the beam radius in the waist by level $1/e^2$, n is the BWO refraction index, λ is the wavelength) to the crystal length at 20, 40 and 90 mm focal distances were equal to 0.02, 0.08 and 0.41, respectively. Thus, as in [4–7], the crystal length with all focuses did not limited the most intense portion of the beam (waist) corresponding to the efficient nonlinear interaction length.

With „soft" focusing ($f = 90$ mm), generation efficiency was $\sim 1\%$. For focusing with 40 mm lens, the highest conversion efficiency (10%) was achieved. For focusing with 20 mm lens system, the efficiency reached 5% with energy $2.5\,\mu$J, while no efficiency drop was observed with energy increase. Therefore, conversion efficiency could be probably higher with focusing $f = 20$ mm, but the used laser system did not allow to implement the mode with higher energy.

Focus position influence

In [4], SRS generation efficiency of mode ν_1 achieved 20% with focusing using a 35 mm lens, which is two times higher than the efficiency achieved by us at $z = 0$. Due to the proximity of focal distances in the used lens, this difference may be attributed to another factor, for example, to the linear focus position inside the sample, which was not specified in [4]. In order to study the influence of this factor, dependence of SRS generation efficiency on the linear focus position inside the crystal

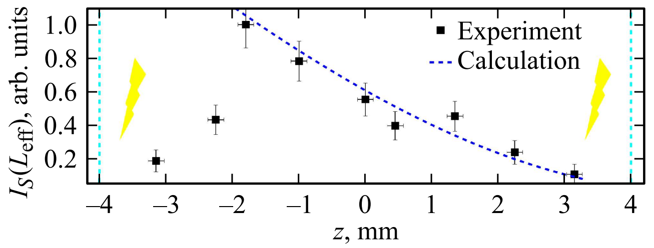

Figure 3. Dependence of the strongest SRS mode amplitude on the linear focus position inside the crystal. Dashes — calculation of conversion efficiency depending on the linear focus position taking into account varying filament length.

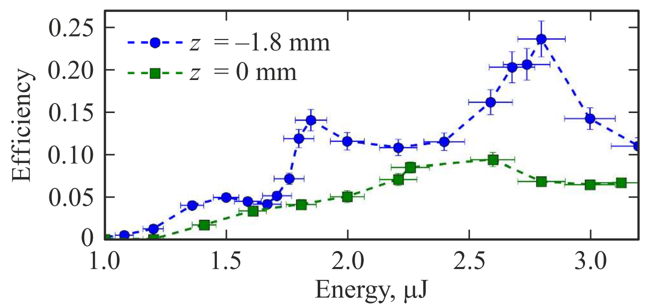

Figure 4. Dependence of generation efficiency of mode 925 cm^{-1} for two positions ($z = 0$ mm and $z = -1.8$ mm) on the laser pulse energy.

was measured. By displacing the crystal along the focused beam from the initial position ($z = 0$) at the pulse energy of 2.6 μJ (maximum generation efficiency with 40 mm lens focusing), the strongest SRS mode amplitude was measured by a spectrometer depending on the linear focus position (Figure 3). In positions marked by „lightning", plasma plume occurred indicating the damage of the front or rear crystal face.

When the linear focus moved from the crystal center to its front face, SRS line amplitude was reduced, and when the linear focus moved to the rear face, the maximum was achieved at $z \approx -1.8$ mm, and when the focus moved further to the rear crystal face, sharp decrease of SRS mode amplitude was observed.

Dependences of the conversion efficiency on the laser pulse energy for two positions (linear focus in the crystal center ($z = 0$) and in the position of the maximum amplitude of the strongest SRS mode ($z = -1.8$ mm)) are shown in Figure 4. The maximum generation efficiency of SRS component SRS component with a shift of 925 cm^{-1} was equal to 23%, which correlates with the results in [4]. It should be noted that the linear focus position shift resulted in the change in the best laser pulse energy at which the maximum SRS conversion efficiency is achieved. Thus, in case of shift from $z = 0$ mm to $z = -1.8$ mm, optimum energy of laser pulse increased from 2.6 μJ to 2.8 μJ.

Analysis and discussion of findings

The obtained experimental data cannot be explained in the approximation where the interaction region is defined only by the geometric optics. In case of linear propagation of the laser pulse in the crystal (only beam size variations due to the lens focusing are taken into account), the highest SRS conversion efficiency is achieved at $z = 0$ mm. Since the ratio of Rayleigh length to the crystal length is 0.08 by focusing with 40 mm lens, when the linear focus is shfited by 1.8 mm towards the rear face, the efficiency shall decrease negligibly (by $\sim 3\%$). Thus, when addressing the constructive interference of SRS and SPM in BWO crystal, self-focusing effect shall be also taken into account.

Critical self-focusing power P_{cr} of the laser pulse is calculated using expression [8]

$$P_{\text{cr}} = 3.77\lambda^2/(8\pi n_0 n_2), \tag{1}$$

where n_0, n_2 are the linear and nonlinear refraction indices. For BWO crystal, $n_0(515\,\text{nm}) = 1.85$ [9]. Nonlinear refraction index of BWO is $n_2 = 6.4 \cdot 10^{-15}\,\text{cm}^{-2}\text{W}$, however, its Kerr nonlinearity is noninstantaneous with a decay time of 0.35 ps compared with the laser pulse length [6,7]. Therefore, to address the self-focusing effect, we used effective nonlinear refraction index $n_2 = 1.7 \cdot 10^{-15}\,\text{cm}^{-2}\text{W}$ measured using Z-scan technique [10], i.e. the technique based on the change in the spatial shape of the beam due to self-focusing, when the response time is not taken into account.

Using expression (1), we found that the critical self-focusing power for our conditions was $P_{\text{cr}} = 0.13$ MW, which corresponds to laser pulse energy $E = 40$ nJ. Therefore, efficient SRS component generation with 925 cm^{-1} shift is accomplished using a laser pulse power which is much higher than the critical power, and the self-focusing effect has a considerable impact on the interaction geometry. For a collimated beam, the nonlinear focus position is

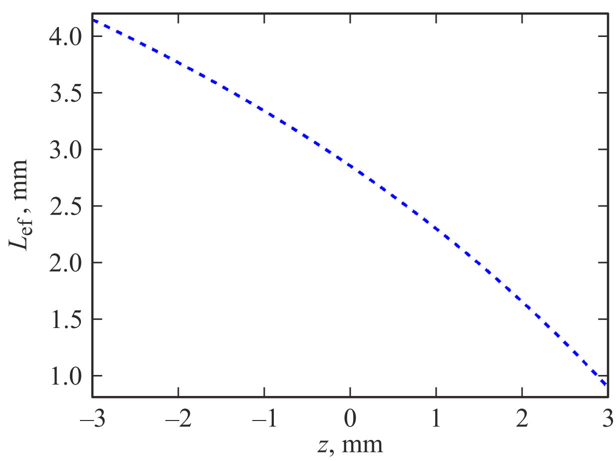

Figure 5. Calculated filament length from the linear focus position inside a 8 mm long BWO crystal with laser pulse energy 2.6 μJ.

determined using Marburger semiempirical formula [11]:

$$z_{sf} = \frac{0.367ka^2}{\sqrt{(\sqrt{P/P_{\mathrm{cr}}} - 0.852)^2 - 0.0219}},$$

where k is the wave number, a is the beam radius at level $1/e$, P is the peak emission power, P_{cr} is the critical self-focusing power. When there is additional focusing with a lens with focal distance f, the distance to nonlinear focus z_{fil} is calculated using the following equation

$$\frac{1}{z_{\mathrm{fil}}} = \frac{1}{z_{sf}} + \frac{1}{f}. \qquad (2)$$

It should be noted that self-focusing only occurs in BWO sample, since the critical self-focusing power in air ($P_{\mathrm{cr}}(\mathrm{air}) \sim 10^9\,\mathrm{W}$ [12] corresponds to laser pulse energy of 0.4 mJ) is considerably higher than pulse energy/power in our experiment. Therefore in our case, f in equation (2) corresponds to the equivalent focal distance of the lens on the front crystal face.

Linear focus displacement from the front to rear crystal face results in the increase in nonlinear phase shift and, thus, to an earlier beam collapse (filament start). According to the moving focuses model, the filament end is determined by the linear focus because the nonlinear focus corresponds to the convergence of the central beam portion (the most intense portion) with nonlinear focuses of less intense beam portions located between the nonlinear focus and linear focus. In this case, the filament length may be assessed as a difference of distances from the linear to nonlinear focuses, $L_{\mathrm{ef}} = F - z_{\mathrm{fil}}$. The curve of filament length vs. linear focus position at pulse energy of $2.6\,\mu\mathrm{J}$ is shown in Figure 5.

With constant laser pulse energy and unchanged lens, the emission intensity inside the filament may be assumed as constant („intensity clamping" effect) [13]. With linear focus displacement, the filament length increases, i.e. the nonlinear interaction length increases. Taking into account the exponential growth of SRS component radiation intensity in transient SRS regime [14]

$$I_S(L) = I_S(0)\exp(\sqrt{8gI_pt_pL_{\mathrm{eff}}/T_2}),$$

where $I_s(0)$ and $I_s(L)$ are the SRS emission intensities at the SRS medium entrance and exit, g is the SRS amplification coefficient, I_p is the pumping intensity, t_p is a pulse duration, T_2 is a dephasing time of the medium, L_{eff} is the efficient SRS interaction length, calculation of SRS conversion from the linear focus position has shown good correlation with the experiment for $z \geq -1.8\,\mathrm{mm}$. However, with further shift of the linear focus towards the rear face ($z < -1.8\,\mathrm{mm}$), the experiment showed sharp decrease of the stokes component amplitude, which is not explained in the presented model. This efficiency drop may be attributed to: 1) mismatching of group velocities of SRS pulses and pumping equal to 50 fs/mm for our conditions, 2) damage of the rear crystal face without bright plasma plume, 3) retaining the intense radiation portion behind the linear focus of the laser beam (post-filamentation channel [14]), which may contribute to the SRS amplification, but in case of linear focus displacement towards the rear crystal face, the post-filamentation channel can be limited by the crystal dimensions.

Conclusion

Effect of constructive interference of SRS and SPM effects of 0.3 ps 515-nm laser pulses in BWO crystal was experimentally studied with varying focusing conditions. With „soft" focusing ($f = 90\,\mathrm{mm}$), generation efficiency of SRS peak $\nu_1 = 925\,\mathrm{cm}^{-1}$ was $\sim 1\%$. With harder focusing ($f = 40$ and $20\,\mathrm{mm}$) and other conditions being equal, the generation efficiency of this peak achieved 10% and 5%, respectively.

It has been found that, when the linear pumping emission focus moved from the front to rear face of BWO crystal, exponential growth of SRS peak amplitude ν_1 is observed. Maximum conversion efficiency achieved 23%, when the linear focus was displaced from the center by $\sim 2\,\mathrm{mm}$ to the rear face of 8-mm crystal specimen. However, with further displacement of the linear focus to the rear face (at a distance less than 2 mm from the face), the peak amplitude decreased dramatically.

The assessments have shown that the exponential growth was due to the presence of nonlinear (self-)focusing. Linear focus displacement to the rear crystal face results in additional nonlinear phase shift and, accordingly, in the increase in the distance between the linear and nonlinear focuses (filament length increase). Consequently, effective length of the nonlinear interaction (SRS amplification length) increases and the SRS peak amplitude grows exponentially. When the linear focus approached the rear face at a distance less than 2 mm, sharp drop of the SRS conversion efficiency was observed. Various effects that may be associated with this were suggested.

Funding

This study was supported by grant No. 22-79-10068 provided by the Russian Science Foundation (https://rscf.ru/project/22-79-10068/).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] A.A. Ionin, I.O. Kinyaevsky, Yu.M. Klimachev, A.Yu. Kozlov, A.A. Kotkov, O.A. Rulev, A.M. Sagitova, L.V. Seleznev, D.V. Sinitsyn. Zhurn. prikl. spektr., **89** (4), 443 (2022) (in Russian). DOI: 10.47612/0514-7506-2022-89-4-443-454

[2] V.S. Gorelik. Sibirskiy fizicheskiy zhurnal, **13** (3) 62 (2018). (in Russian). DOI: 10.25205/2541-9447-2018-13-3-62-68

[3] A. Dubietis, G. Tamošauskas, R. Šuminas, V. Jukna, A. Couairon. Lithuanian J. Physics, **57** (3), 13 (2017). DOI: 10.3952/physics.v57i3.3541
[4] I.O. Kinyaevskiy, V.I. Kovalev, P.A. Danilov, N.A. Smirnov, S.I. Kudryashov, L.V. Seleznev, E.E. Dunaeva, A.A. Ionin. Opt. Lett., **45** (8) 2160 (2020). DOI: 10.1364/OL.391550
[5] I. Kinyaevskiy, V. Kovalev, P. Danilov, N. Smirnov, S. Kudryashov, A. Koribut, A. Ionin. Chinese Opt. Lett., **21** (3) (2023). DOI: 10.3788/COL202321.031902
[6] I. Kinyaevskiy, V. Kovalev, P. Danilov, N. Smirnov, S. Kudryashov, A. Koribut, A. Ionin. Opt. Lett., **46** (3), 697 (2021). DOI: 10.1364/OL.417661
[7] I.O. Kinyaevskiy, V.I. Kovalev, A.V. Koribut, P.A. Danilov, N.A. Smirnov, S.I. Kudryashov, Ya.V. Grudtsyn, E.E. Dunaeva, V.A. Trofimov, A.A. Ionin. J. Rus. Laser Research, **43**, 315 (2022). DOI: 10.1007/s10946-022-10053-2
[8] R.Y. Chiao, E. Garmire, C.H. Townes. Phys. Rev. Lett., **13** (15), 479 (1964). DOI: 10.1103/PhysRevLett.13.479
[9] L.I. Ivleva, I.S. Voronina, P.A. Lykov, L.Y. Berezovskaya, V.V. Osiko. J. Crystal Growth, **304** (1), 108 (2007). DOI: 10.1016/j.jcrysgro.2007.02.020
[10] A.I. Vodchits, V.A. Orlovich, P.A. Apanasevich, T.T. Basiev, P.G. Zverev. Opt. Mater., **29** (12), 1616 (2007). DOI: 10.1016/j.optmat.2006.08.005
[11] J.H. Marburger. Progress in Quantum Electronics, **4**, 35 (1975). DOI: 10.1016/0079-6727(75)90003-8
[12] E.T.J. Nibbering, G. Grillon, M.A. Franco, B.S. Prade, A. Mysyrowicz. JOSA B, **14** (3), 650 (1997). DOI: 10.1364/JOSAB.14.000650
[13] V.P. Kandidov, V.Yu. Fedorov, O.V. Tverskoi, O.G. Kosareva, S.L. Chin. Quant. Electron., **41** (4) 382 (2011). DOI: 10.1070/QE2011v041n04ABEH014486.
[14] A. Penzkofer, A. Laubereau, W. Kaiser. Prog. Quant. Electron., **6** (2), 55 (1979). DOI: 10.1016/0079-6727(79)90011-9
[15] Y. Geints, O. Minina, A. Zemlyanov. JOSA B, **39** (6), 1549 (2022). DOI: 10.1364/JOSAB.453694

Translated by E.Ilyinskaya