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Stimulated Raman scattering of titanium-sapphire laser pulses with duration from 7 ps to 45 ps in $BaWO_4$ crysral

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Transient stimulated Raman scattering of titanium-sapphire laser pulses with varying duration in a $BaWO_4$ crystal was experimentally studied at its dominant mode $\nu_1 = 925\,\text{cm}^{-1}$ with the dephasing time $T_2 \sim 6.6$ ps. For pulses with a duration of 7−17 ps (corresponding to $\sim T_2$ — $2.5T_2$), the maximum conversion efficiency was ∼ 1%, which was achieved at the same laser pulse fluence of $1.2 \pm 0.2\,\text{J/cm}^2$. At a pulse duration of 45 ps ($\sim 7T_2$), the SRS conversion efficiency was 7 times higher (∼ 7%), and the SRS „threshold" fluence was 2 times lower. It has been shown that to reduce the threshold and to increase the efficiency of stimulated Raman scattering, the femtosecond pulse should be stretched in time at least to $\sim 7T_2$.

Keywords: stimulated Raman scattering, $BaWO_4$, chirped pulses, transient regime.

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Introduction

Stimulated Raman scattering (SRS) is a widely used method for varying the wavelength of laser radiation [1] whose efficiency may achieve almost quantum limit [2]. However, for ultrashort (femtosecond) laser pulses that are comparable or shorter than the oscillation dephasing time T_2, SRS efficiency is much lower due to the transient regime of this interaction process [3]. Accordingly, high intensity of pumping radiation is required to increase the SRS efficiency for ultrashort laser pulses. However, high intensity, in turn, enhances the effect of other nonlinear effects such as self-phase modulation, nonlinear absorption, etc. [4–7]. To solve this problem, a technique is used where the pumping pulse is extended in time up to a value higher than T_2 using, for example, diffraction gratings.

Several studies are known [7–12] that demonstrate high, up to 40% [11], SRS efficiency of chirped laser pulses. Despite the success in this area, the problem of increasing SRS efficiency by controlling the pumping pulse parameters has been solved incompletely. In particular, optimization of chirp (pulse width) quantity has not been addressed in detail. The problem is in the fact that, if the chirped pulse width is lower or comparable with T_2 (small chirp), then, as mentioned above, the effective SRS amplification factor will be lower due to transient regime of the process. If the pulse width is too long (large chirp), this will result in a decrease in effective SRS amplification factor due to fast carrier frequency shift of the pumping pulse [3,10]. It should be noted that pulse width also affects other factors, for example, optical damage threshold, that also define the maximum attainable SRS efficiency. Therefore the purpose of this study was to determine the SRS efficiency of chirped laser pulses of femtosecond titanium-sapphire laser with varying pulse width (chirp quantity). The study uses pulses with positive chirp that initially was set by a „stretcher" built in the titanium-sapphire laser. For this studies, $BaWO_4$ (BWO) crystal was chosen as one the most effective SRS converters of pumping frequency [2,13] that had been tested numerous times in experiments with chirped laser pulses [8,10].

Experiment setup

Experimental optical setup is shown in Figure 1. The experiment used Start 480M titanium-sapphire laser system (Avesta Proekt, Russia): central wavelength $0.75\,\mu$m, spectrum full width at half maximum 10 nm ($\sim 150\,\text{cm}^{-1}$), spectrally limited pulse width 100 fs, energy up to 6 mJ. Radiation was output, by-passing the built-in laser system compressor, and send to a self-made controlled compressor consisting of two diffraction gratings 1200 groves/mm and back reflector. The time-compressed pulse was fed either to GPI PS-1/S1 „streak"-camera (Institute of General Physics of Academy of Sciences, Russia) to measure its width or to the SRS converter. The SRS converter consisted of a lens with a focal distance of 35 cm and $BaWO_4$ crystal 8 mm in length. The crystal was placed after the laser beam focus at 37 cm from the lens, crystal position was not changed throughout the experiment. It should be noted that at the crystal shift to radiation focus, crystal damage was observed independently of the pulse width. The radiation coming from the SRS crystal was fed to ASP-150 spectrometer (Avesta Proekt, Russia). To change the laser pulse energy, a diffraction attenuator was installed before the compressor.

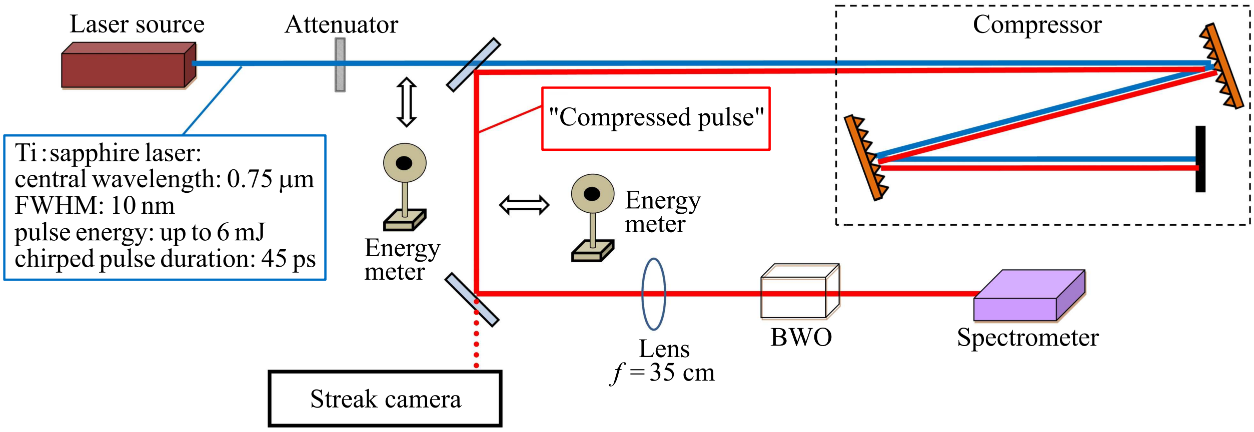

Figure 1. Experimental optical setup.

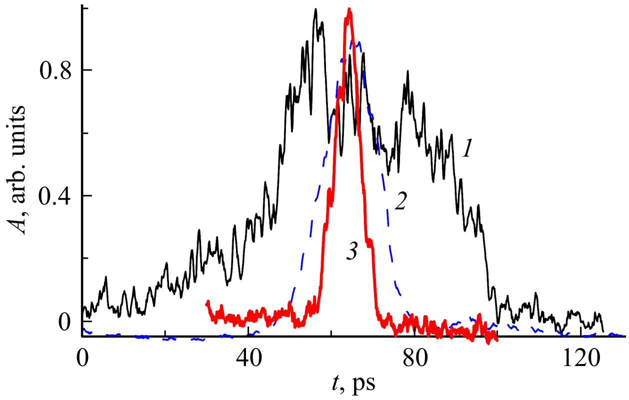

Figure 2. Temporal pulse shapes before the compressor (*1*) and after the compressor with different spacing between diffraction gratings: $t_p = 17$ (*2*), 7 ps (*3*).

The test sample of BWO crystal 8 mm in length was grown from platinum crucibles in air by the Czochralski method at Prokhorov Institute of General Phhysics RAS [14]. This material features high steady-state SRS amplification factor 8.0 ± 1.6 cm/GW ($\lambda = 1.06\,\mu$m) at mode $\sim 925\,\mathrm{cm}^{-1}$ for which dephasing time T_2 is 6.6 ps [15]. The SRS conversion efficiency was increased using a seed by a broadband nanosecond pulse representing amplified spontaneous emission of a titanium-sapphire laser multipass amplifier and propagating together with the laser pulse [8,16].

Experimental results

Positively chirped laser pulse width (without compressor) was $t_p = 45 \pm 5$ ps at half maximum (curve *1* in Figure 2). Minimum spacing between the gratings in the compressor was $L = 17$ cm and was limited by laser beam „cutoff" by optical elements included in the compressor. At $L = 17$ cm, the pulse width was $t_p = 17 \pm 2$ ps (curve *2* in Figure 2). Increase in grating spacing controlled pulse width, $t_p = 10 \pm 1$ ps and 7 ± 1 ps pulses were obtained (curve *3* in Figure 2). It should be noted that at the laser pulse energy at the compressor entrance of ~ 6 mJ, the laser pulse energy after the compressor was ~ 1 mJ due to low ($\sim 70\%$) reflectance of diffraction gratings.

Laser pulses with various widths ($t_p = 7{-}45$ ps) and an energy of 1 mJ was directed to the SRS converter whose spectrum was recorded by a spectrometer. Figure 3 shows laser pulse spectra after the SRS converter measured at $t_p = 7$ (*a*), 10 (*b*), 17 (*c*) and 45 ps (*d*). These widths corresponded approximately to T_2, $1.5T_2$, $2.5T_2$ and $7T_2$, and the chirp defined in the linear case as ratio of spectrum width to pulse width, $\Delta f/\Delta t$, was equal to 0.7, 0.5, 0.3, 0.1 ps^{-2}, respectively. Spectra in Figure 3 were normalized to amplitude of the transmitted pumping spectrum.

For 7–17 ps laser pulses, that corresponded approximately to $T_2{-}2.5T_2$, SRS conversion efficiency was almost the same: $(1 \pm 0.5)\%$. Efficiency was calculated as area under SRS peak devided on area under total spectrum. At $t_p = 45$ ps, the SRS peak amplitude was much higher and achieved $\sim 20\%$ of the transmitted pumping spectrum amplitude, and the efficiency was $\sim 7\%$. It should be noted that at $t_p = 45$ ps strong change in the pumping pulse spectrum shape was observed that may be caused by collinear four-wave mixing as has been previously observed in [17].

Figure 4, *a* shows the SRS generation „threshold" (SRS efficiency — 1%) in energy density units vs. laser pulse width as found from the experiment. SRS generation „threshold" for relatively short chirped pulses ($t_p = 7{-}17$ ps, $T_2{-}2.5T_2$) was at approximately the same pumping pulse density $1.2 \pm 0.2\,\mathrm{J/cm}^2$ that is typical for transient SRS [1,3]. At $t_p = 45$ ps, „threshold " SRS energy density reduced at half (to $\sim 0.6\,\mathrm{J/cm}^2$) that indicates a transition to the steady-state SRS mode. The „threshold" SRS intensity for $t_p = 45$ ps was $\sim 13\,\mathrm{GW/cm}^2$ which is ~ 12 times lower then for $t_p = 7$ ps.

Black points in Figure 4, *b* show exponential gain increment $G = Igl$ for the SRS „threshold" vs. rel-

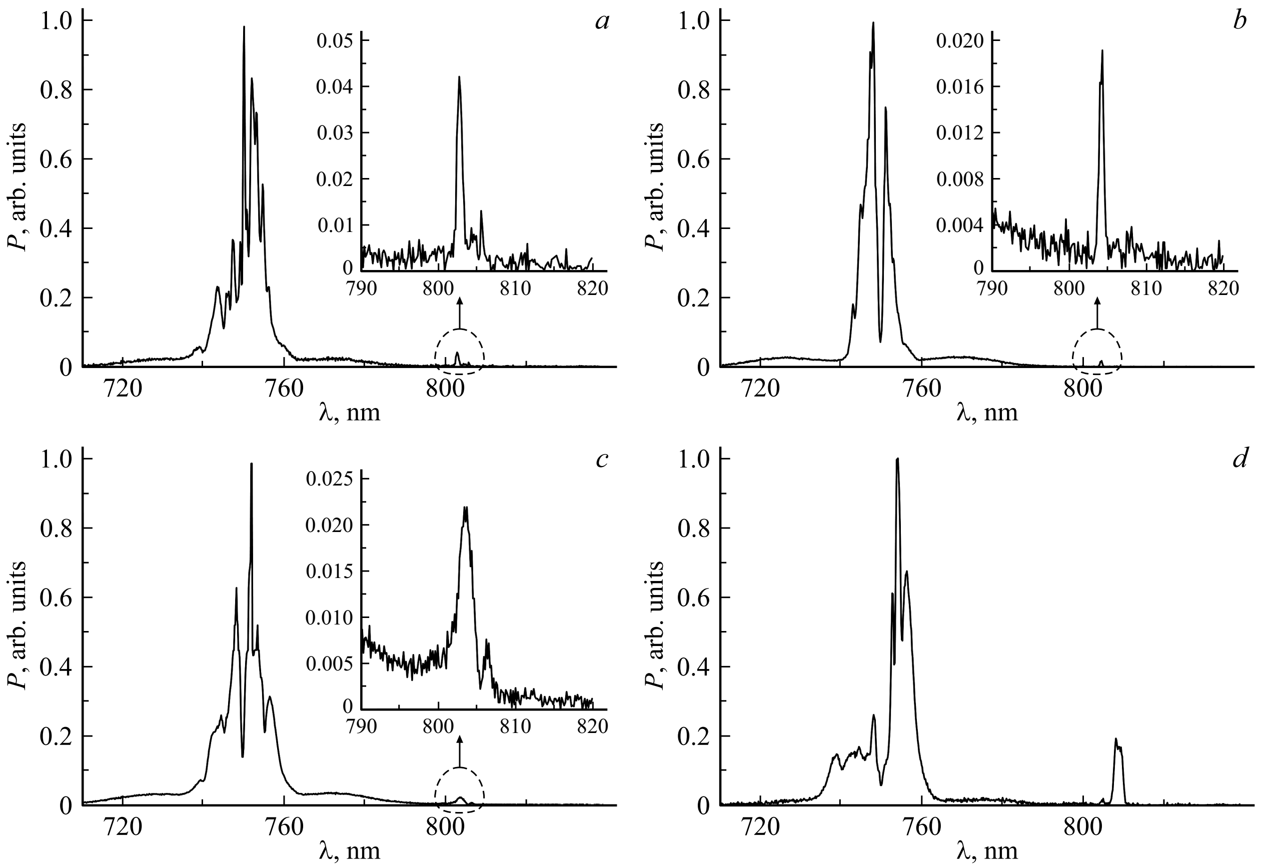

Figure 3. Radiation spectra passed through BWO crystal, normalized to unity, for 7 (*a*), 10 (*b*), 17 (*c*) and 45 ps laser pulses (*d*).

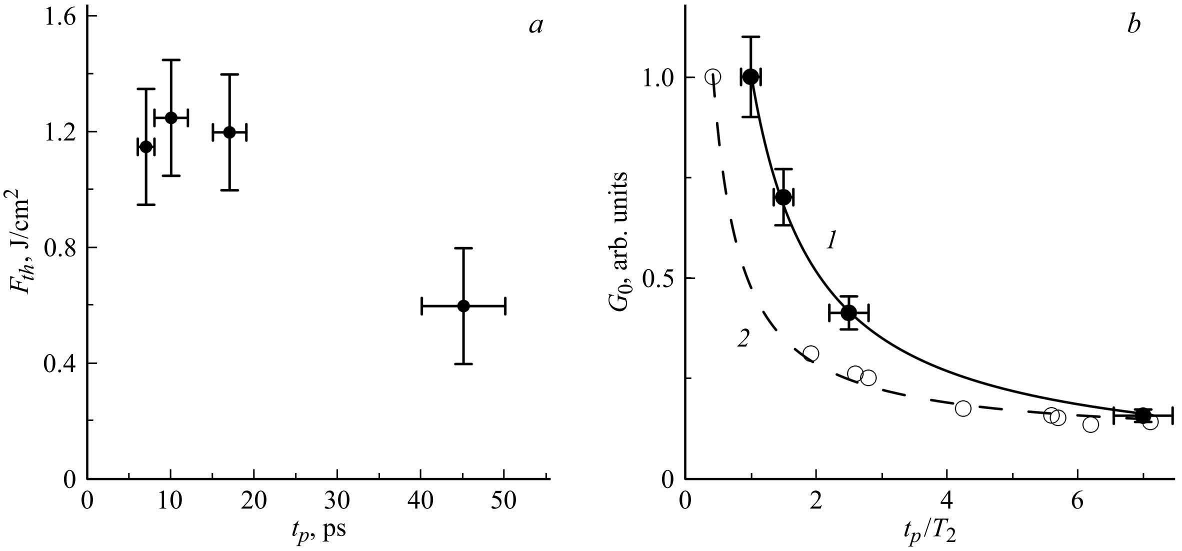

Figure 4. (*a*) „SRS generation threshold“ depending on the pumping pulse width in energy density units; (*b*) dependence of the gain increment $G = Igl$ for SRS generation „threshold“ on relative pulse width t_p/T_2: *1* — this study, *2* — data from [18].

ative pulse width t_p/T_2. Circles in Figure 4, *b* also show the findings of [18]. For ease of analysis, the dependences in Figure 4, *b* are normalized. It should be noted that the experiment in [18] was carried out with spectrally limited 11 ps, laser pulses, and the relative pulse width t_p/T_2 varied by using various SRS crystals at T_2 from 1.18 ps for TeO_2 to 26.5 ps for $Ba(NO_3)_2$. Experimental points were interpolated by function $G = k_1 + k_2/(t_p/T_2)$ [19], where k_1 and k_2 are interpolation factors.

Dependences in Figure 4, *b* are in reasonable agreement with each other, however, for chirped pulses (curve *1*), G grows faster with the decrease in the relative pulse width. This may be caused by the growth of contribution of other nonlinear effects — nonlinear absorption [10] or four-wave interaction [17] that are more clearly manifested for shorter pulses with higher intensity.

Findings and conclusion

$BaWO_4$ crystal was used for experimental study of SRS pulses of the femtosecond titanium-sapphire laser whose width is varied by the optical compressor. For 7−17 ps (corresponds to $T_2-2.5T_2$) pulses, the maximum conversion efficiency was at 1% level, while the SRS generation „threshold“ was achieved at the same energy density1.2−0.2 J/cm^2. At a pulse width of 45 ps, the SRS conversion efficiency was 7 times higher and the SRS generation threshold in the energy density and intensity units decreased to ~ 0.6 J/cm^2 and 13 GW/cm^2, respectively. SRS „threshold“ behavior during variation of the relative pulse width t_p/T_2 for the chirped pulse, looked the same as for the spectrally limited pulse. To assess the effect of pumping pulse chirping on SRS, experiments with high pulse width (chirp) range and also with different sign chip are required. However, this study suggests that to reduce the threshold and increase the SRS conversion efficiency, the femtosecond laser pulse shall be extended in time at least to $\sim 7T_2$.

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Conflict of interest

The authors declare that they have no conflict of interest.

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