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Spectral Broadening of Chirped Laser Pulse Caused by Four-Wave Mixing in $BaWO_4$ Crystals

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Abstract: The spectral broadening of a chirped, temporally stretched (up to 200 ps) Ti:sapphire laser pulse propagating together with a broadband low-power seed pulse through $BaWO_4$ crystals was experimentally and numerically studied. Our analysis demonstrated that self-phase modulation could not have affected the pulse spectrum under the experimental conditions. The spectral broadening of the Ti:sapphire laser pulse was found to be due to four-wave mixing, which resulted in transferring the power of the central wavelength to the spectral wings. To the best of our knowledge, this is the first demonstration of such origin of laser pulse spectral broadening in crystals.

Keywords: ultrashort laser pulse; chirped pulse; four-wave mixing; spectral broadening; $BaWO_4$ crystal

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1. Introduction

Different nonlinear optical effects related to third-order nonlinearity can occur in a medium in which a high-intensity ultrashort laser pulse propagates, such as third-harmonic generation, stimulated Raman scattering (SRS), self-focusing, four-wave mixing (FWM), self-phase modulation (SPM), etc. [1]. To eliminate undesirable nonlinear effects such as SPM and self-focusing, ultrashort laser pulses are stretched out in time prior to the required interaction and are afterwards compressed to their transform-limited duration. Chirped (stretched out in time) laser pulses are used in laser amplifiers [2], optical parametric amplifiers [3,4], optical amplifiers/converters based on the SRS effect [5–7], and optical schemes combining several nonlinear effects [8]. Moreover, an ultrashort laser pulse can experience significant temporal stretching just due to the dispersion of the propagation medium.

In experiments on chirped laser pulse SRS in $BaWO_4$(BWO) crystals [6–8], notable spectral broadening of a pump pulse was observed. Usually, the major effect that causes the spectral broadening of a femto–picosecond laser pulse is SPM; however, under those experimental conditions [6–8], the SPM effect was insignificant. Moreover, the origin of chirped pulse spectral broadening was unclear. In this research, we studied this effect and found out that a chirped, temporally stretched Ti:sapphire laser pulse was spectrally broadened in $BaWO_4$ crystals due to a quite different nonlinear effect, namely, FWM, which caused pump pulse spectrum wing enhancement. It should be noted that, to the best of our knowledge, this is the first demonstration of laser pulse spectral broadening in crystals of such origin, although previously, similar effects have been observed in optical fibers [9,10].

2. Experimental Setup and Results

The experiment was carried out with the Ti:sapphire front end of a hybrid Ti:sapphire/KrF laser facility [11]. The optical scheme of the experiment (Figure 1) was close to one of the experiments in [7], but the current research was focused on pump pulse spectral broadening.

A chirped pulse of ~200 ps duration, with the transform-limited laser pulse duration being 90 fs, was taken after the Ti:Sapphire amplifier just before the laser facility compressor. The laser pulse energy was up to 8 mJ and was controlled using a diffraction attenuator.

The chirped laser pulse was temporally and spatially coupled with a spectral broadband (0.6–0.9 μm) seed pulse of 10 ns duration and 2 mJ energy. The seed pulse originated from superluminescence–spontaneous emission enhanced in the multi-pass Ti:Sapphire laser amplifier and was generated concurrently with the laser pulse. Since the seed and laser pulses were spatially and temporally coupled, their energy ratio was not affected by the attenuator. It should be noted that the application of broadband seed radiation is usual practice in nonlinear optics, e.g., in optical parametric amplifiers [3,4]. The radiation spectrum containing the main and seed pulses is presented in the insert of Figure 1, where the spectra of the corresponding pulses approximated by a Gaussian profile are indicated by dashed and dotted lines. The beam cross-section (both pump and seed) had a Gaussian profile with a diameter of 8 mm at the $1/e^2$ level.

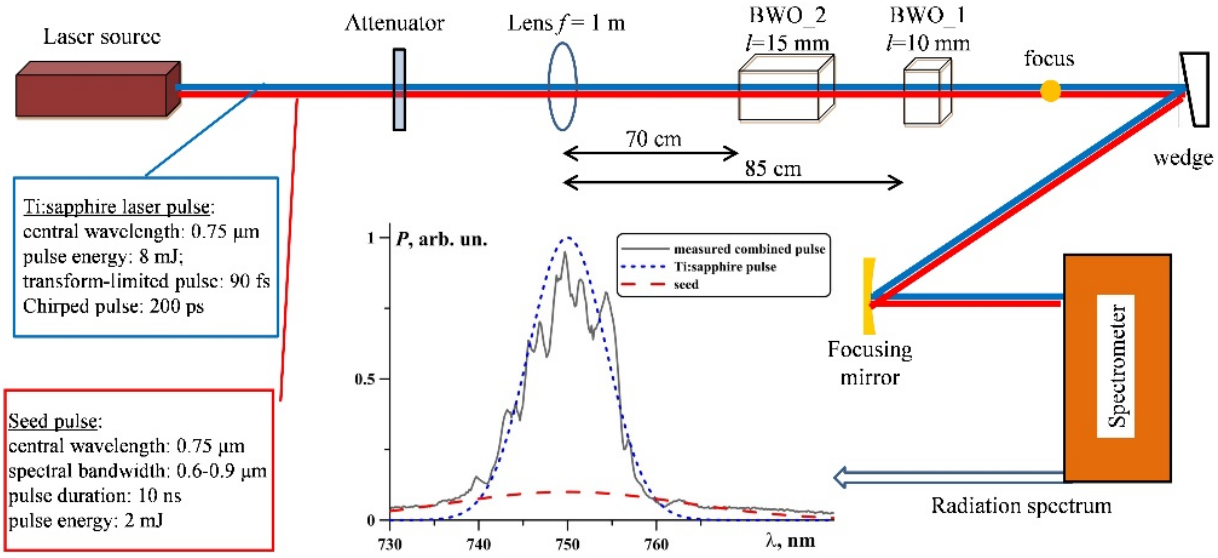

Figure 1. Simplified optical scheme of the experiment. The insert demonstrates the pump pulse spectrum (solid line) combining the spectra of the laser pulse (dotted line) and seed pulse (dashed line).

The laser pulse was directed through a lens with the focal length of 1 m into a BWO crystal (BWO_1) of 10 mm in length. The BWO crystal was located in front of the focus of the laser beam at a distance slightly shifted from the position where crystal optical damage appeared (the distance of 85 cm from the focusing lens). To enhance the spectral broadening, the second BWO crystal sample (BWO_2) of 15 mm in length was installed before the BWO_1 sample at the distance of 70 cm from the focusing lens. The laser beam linear polarization was perpendicular to the optical c-axis of the BWO samples. The BWO crystals were grown and manufactured at General Physics Institute of RAS.

Behind the BWO crystals, the laser beam was focused in air; the divergent laser beam after the focus was attenuated with a couple of wedge plates (indicated as a single wedge in Figure 1 for simplicity) and focused into an ASP-480 spectrometer (Avesta Project Ltd., Moscow, Russia) with a spherical mirror. It should be noted that the focusing in air did not cause any nonlinear effects because the laser pulse power of 40 MW (8 mJ/0.2 ns) was significantly lower than the self-focusing critical power of 2.4 GW. It was verified in the experiment by removing the BWO crystals.

The spectrum of the initial pulse had a central wavelength of 0.75 μm, with the spectral bandwidths being between ~10 nm and ~15 nm at the $\frac{1}{2}$ and 1/e levels, respectively (see line 1 in Figure 2a). After passing through the first BWO crystal, the spectrum of the laser pulse (at 8 mJ energy) was deformed and slightly broadened (line 2 in Figure 2a). The spectral broadening after passing through the two BWO samples became more pronounced and stronger (line 3 in Figure 2a).

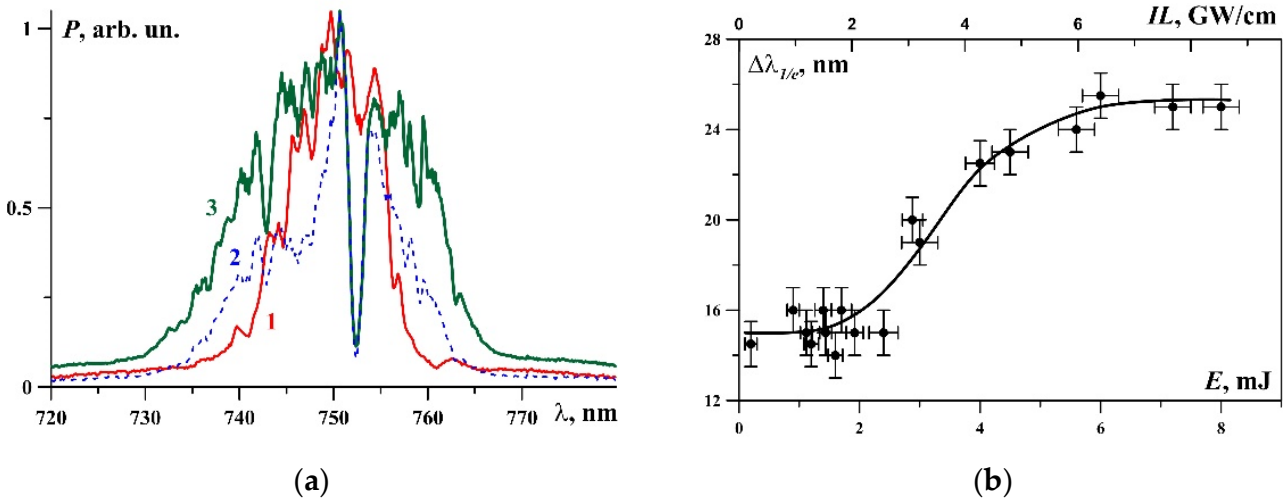

Figure 2. The spectrum of the laser pulse before (1) and after passing through one (2) and two (3) BWO crystals (**a**); the spectral bandwidth at the 1/e level of the laser pulse after the BWO crystals vs. pulse energy and parameter *IL* (see below) (**b**).

The dependence of the measured pulse spectrum bandwidth on pump pulse energy is presented in Figure 2b. The pulse bandwidth was about the same within the measurement error at pulse energy below 3 mJ, increased at pulse energy higher than 3 mJ, reached about 25 nm (at 1/e level) at 6 mJ energy, and was about the same at pulse energy above 6 mJ.

3. Numerical Analysis

To analyze the experimental data, the pulse energy was transformed into intensity units. Afterwards, we calculated parameter *IL* (the upper X-axis in Figure 2b), which is the product of radiation intensity by crystal length. Since the experimental scheme comprised two separated BWO crystals, parameter *IL* was calculated using the following expression:

$$IL = I_1 \cdot L_1 + I_2 \cdot L_2 \quad (1)$$

where $I_{1,2}$ are the intensities at the entrance of the first and second BWO crystals, respectively; and $L_{1,2}$ are the lengths of the first and second BWO crystals, respectively. For instance, at 3 mJ laser pulse energy, the radiation intensities at the entrance of the front (BWO_2) and rear (BWO_1) crystals were 0.7 GW/cm^2 and 2.1 GW/cm^2, respectively. The intensity at the entrance of the rear (BWO_1) crystal was higher due to the focused laser beam converging.

To find out the origin of chirped pulse spectral broadening, we carried out a numerical analysis of the possible nonlinear effects. The first effect that is usually responsible for the spectral broadening of ultrashort laser pulses is SPM. The spectral broadening induced by SPM was calculated using the following expression [1]:

$$\Delta\nu_{SPM} = \frac{\Delta\omega}{2\pi \cdot c} = \frac{n_2 \cdot IL}{\lambda \cdot t \cdot c}[cm^{-1}] \quad (2)$$

where n_2 = 6.5 × 10^{-15} cm^2/W [12,13]—nonlinear refractive index of BWO crystals; λ = 0.75 μm—radiation wavelength; t = 200 ps—pulse duration; and c—speed of light. According to Equation (2), the SPM-induced spectral broadening would have been comparable to the initial spectrum (about 12 nm/200 cm^{-1}) at $IL \approx 10^4$ GW/cm, which is three orders of magnitude higher than in the experiment. Thus, SPM could not have considerably affected the laser pulse spectrum.

Another nonlinear effect that can affect the laser pulse spectrum is FWM. It has been previously observed in optical fibers [9,10]. In this case, low-power spectrum wings (anti-

Stokes $\omega_0 + \Delta\omega$ and Stokes $\omega_0 - \Delta\omega$ components) are amplified due to intense emission corresponding to the maximum spectrum at ω_0. The four interacting waves are as follows:

$$\omega_1 = \omega_2 = \omega_0;\ \omega_3 = \omega_0 + \Delta\omega;\ \omega_4 = \omega_0 - \Delta\omega \quad (3)$$

where ω_0—angular frequency corresponding to radiation wavelength λ = 0.75 μm; and $\Delta\omega$—variable frequency shift. It should be noted that the spectrum of the laser pulse after the BWO crystals (Figure 2a) had a dip at the central wavelength. This effect could have taken place because of the transferring of power corresponding to the central wavelength to the spectral wings. This fact indirectly confirmed our hypothesis that the spectral broadening origin was FWM.

In order to make the FWM process efficient, the phase-matching conditions should be met. The phase-matching factor, *PM*, for collinear laser beams, which accounts for reducing the conversion efficiency due to phase mismatching, was calculated using the following expression:

$$PM(\Delta\omega) = sinc^2\left(\frac{\Delta k(\Delta\omega) \cdot L}{2}\right), \quad (4)$$

where L = 25 mm—total length of the two BWO crystals; and $\Delta k(\Delta\omega)$—wave–vector mismatching calculated using the following expression:

$$\Delta k = \frac{\omega_1 \cdot n^0(\omega_1)}{c} + \frac{\omega_2 \cdot n^0(\omega_2)}{c} - \frac{\omega_3 \cdot n^0(\omega_3)}{c} - \frac{\omega_4 \cdot n^0(\omega_4)}{c}. \quad (5)$$

The meanings of the frequencies from ω_1 to ω_4 are presented in Equation (3). The dispersion of the linear refractive index for BWO crystals was taken from [14]. All interacting waves had ordinary polarization because field vector E was perpendicular to the optical c-axis of the BWO crystals. The *PM* factor at variable $\Delta\omega$ covering the wavelength range of 0.72–0.78 μm was calculated and is presented in Figure 3a. The calculation showed that the spectral width of phase matching (PM factor at the $\frac{1}{2}$ level) for the considered FWM process was 25 nm, which coincided with the spectral bandwidth of the laser pulse broadened in BWO crystals.

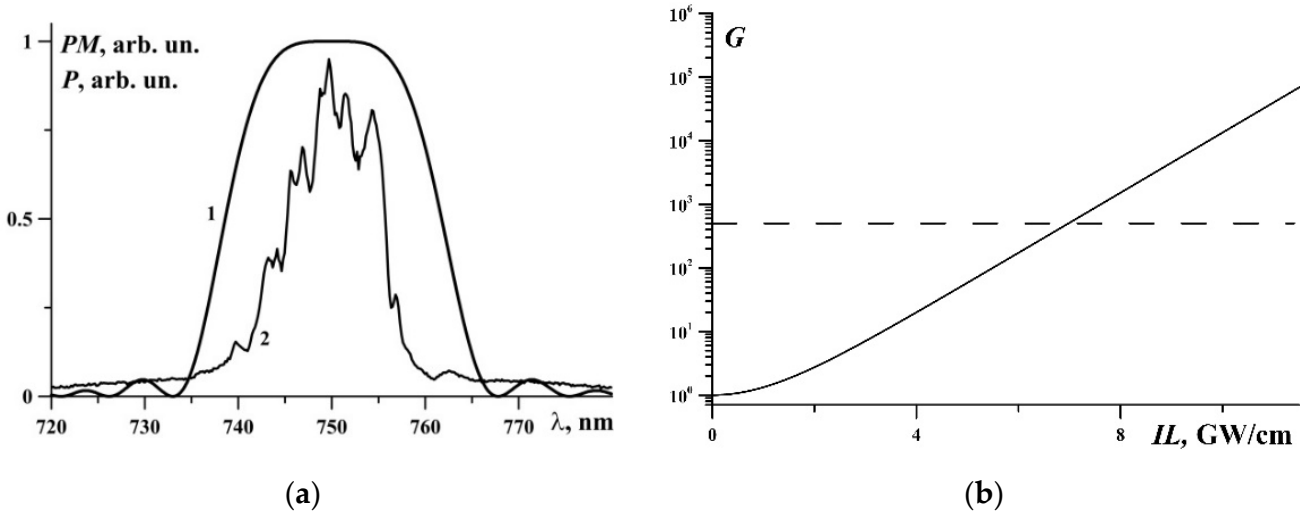

Figure 3. The *PM* factor (1) and pump laser pulse spectrum (2) (**a**); gain ratio *G* versus *IL* parameter, where the dashed line corresponds to the required gain level (**b**).

The next point to verify this model was the evaluation of FWM efficiency through the calculation of a gain ratio *G* that had to be about 5×10^2. The required value of *G* followed on from the following relation between the pump and seed pulse power: The spectral brightness of the seed pulse was about 10% of that of the pump pulse (see insert

in Figure 1); however, the laser pulse duration (200 ps) was 50 times shorter than the seed pulse duration (10 ns). Therefore, the relation between the laser and seed pulse power (required G) was $G = 1/(0.1 \cdot 0.02) = 5 \times 10^2$. It should be noted that the spectral wings of the pump pulse could have provided a higher seeding level for FWM; however, it did not happened because the laser pulse was chirped and its spectral components were separated in time.

The theory of FWM and amplification has been studied considering different media and conditions; see, for example, [1,15]. For simplicity, we considered the following approximation of FWM applicable to our experiment: slowly varying envelopes, because the pulse duration of 200 ps was significantly longer than the optical cycle (~2.5 fs); exact phase matching (see Figure 3a); the difference between the frequencies of the interacting plane waves was neglected ($\Delta\lambda/\lambda$ = 25 nm/750 nm $\approx$ 0.03); no linear nor nonlinear dispersion due to long (~200 ps) laser pulse; no optical losses due to absorption, because the BWO crystal bandgap Eg = 5.26 eV was significantly higher than the photon energy of 1.65 eV. In addition, for simplicity, we considered a non-depleted pump, because if the FWM efficiency is high enough for pump pulse depleting, then it provides the required spectral broadening by default. With these simplifications, a solution to the wave equations for FWM gain G of a Stokes wave can be presented in the following form (see Equation (10.4.7) in [15]):

$$G = \frac{I_3}{I_3^0} = 1 + \sinh^2(\gamma \cdot IL), \tag{6}$$

where γ is a nonlinear parameter defined as

$$\gamma = \frac{2\pi \cdot n_2}{\lambda}. \tag{7}$$

The calculated gain ratio, G, due to FWM versus the IL parameter is presented in Figure 3b. The dashed horizontal line in Figure 3b indicates the required gain level corresponding to the power ratio of the seed and pump pulses. The calculations showed that the required gain, G, of 5×10^2 was achieved at $IL \sim 7$ GW/cm, which was in adequate agreement with the experimental observation (Figure 2b). It should be noted that the intensity–length product in the experiment could have been 10–15% higher, since we did not take into consideration the convergence of the focused laser beam. However, even if it does take place, the calculation indicated that the FWM gain would be quite sufficient for pulse spectral broadening. Thus, the coincidence of both the calculated phase-matching bandwidth and gain ratio G with the experimental results provides convincing confirmation that the origin of the spectral broadening of chirped laser pulses in BWO crystals is just related to FWM.

4. Conclusions

The unusual spectral broadening of a chirped, temporally stretched (up to 200 ps) Ti:sapphire laser pulse propagating together with a broadband low-power seed pulse through $BaWO_4$ crystals was experimentally and numerically studied. This spectral broadening appeared in the crystal at an intensity–length product above 3 GW/cm. The laser pulse spectral bandwidth reached about 25 nm (at the 1/e level) at 6 mJ energy and was about the same at pulse energy above 6 mJ. It was shown that the origin of this effect was related to four-wave mixing, whereby low-power spectrum wings were enhanced due to powerful radiation at the central wavelength. This effect can reduce the conversion efficiency of nonlinear optical devices using a broadband seed under chirped pulse amplification, and accordingly, it should be taken into account. In addition, we would like to note that similar spectral broadening of chirped laser pulse in BaWO4 crystals can appear without external broadband seed [6]. In this case, a seed could be a spectral component of

the main laser pulse scattered from optical elements or have other nature, which will be studied in our future work.

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