Filamentation-assisted fourth-order nonlinear process in KTP crystal*

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(Received 13 April 2009; revised manuscript received 14 September 2009)

We present an experimental investigation of a filamentation-assisted fourth-order nonlinear optical process in KTP crystals pumped by intense 1.53 eV (807 nm) femtosecond laser pulses. Femtosecond light pulses at 2.58 eV (480 nm) are generated by the fourth-order nonlinear polarization ($P^{(4)}(\omega_2) = \chi^{(4)}(\omega_2, \omega, \omega, \omega, -\omega_1)E^3(\omega)E^*(\omega_1)$, where $E(\omega)$ corresponds to the pump frequency and $E(\omega_1)$ to the supercontinuum generated through filamentation). If the system is seeded by a laser beam at ω_1 or ω_2 and there are spatial and temporal overlaps with the pump beam, $E(\omega_1)$ and $E(\omega_2)$ are simultaneously amplified. When the intensity of the seed laser beam exceeds a certain intensity threshold, the contribution of $P^{(4)}(\omega) = \chi^{(4)}(\omega, \omega_1, \omega_2, -\omega, -\omega)E(\omega_1)E(\omega_2)(E^*(\omega))^2$ becomes non-negligible, and the amplification weakens. The conversion efficiency from the pump to the signal at 2.58 eV (480 nm) attains to 0.1%.

Keywords: parametric amplification, filamentation, fourth-order nonlinear polarization, KTP crystal

PACC: 4265K, 4270D

The developments of lasers and techniques such as Q-switching and mode locking have brought researchers high intensity laser sources to investigate nonlinear optical processes such as second harmonic generation (SHG), sum frequency generation (SFG), different-frequency generation (DFG), optical parametric oscillation (OPO), optical parametric amplification (OPA) processes, etc. More nonlinear phenomena such as supercontinuum generation (SCG)[1−5] and filamentation[3,6,7] were observed as more intense femtosecond laser pulses became available. For a quadratic nonlinear medium such as properly configured KDP[8,9] and BBO crystals,[10,11] SCG and SHG may exist simultaneously. Parametric generation excited by SHG in BBO crystals was predicted[12] and observed.[10] Even though the electric field of the SCG component is two orders of magnitude lower than that of the incident laser,[1] the SCG may still be strong enough to be involved in nonlinear processes, opening the door to a deep understanding of more interesting phenomena.

In Ref. [9], the sum frequency (SF) of the supercontinuum (SC) and the incident laser was used to generate a spectrum much broader than the SC alone. Recently we observed a parametric amplification process based on a fourth-order polarization[13] in which the SC served as the seed.

In the present paper, more details and further results on this work are provided and the conversion efficiency is improved to 0.1% through the application of two orthogonally polarized pump beams. Furthermore, we also find that when the intensity of the seed light exceeds a certain threshold, the optical energy transfers back from the amplified signal to the pump beam.

The experiment was performed by using an amplified femtosecond Ti:sapphire laser system (Spitfire, Spectra-Physics, USA) tuned to 1.53 eV (807 nm). The pulse duration was 130 fs and the beam diametre was 5 mm. The experiment setup is shown in Fig. 1. The laser beam was focused by a 20 cm achromatic focal length into a 2 mm thick, x-cut (100) KTP crystal. A half wave plate was set in front of focal lens to adjust the polarization of the laser, and a fibre spectrometer was used to record the spectrum of the laser after propagation through the crystal.

*Project supported by the National Basic Research Program, China (Grant No. 2006CB806007), and the National Natural Science Foundation of China (Grant Nos. 10574006, 10634020 and 10821062).

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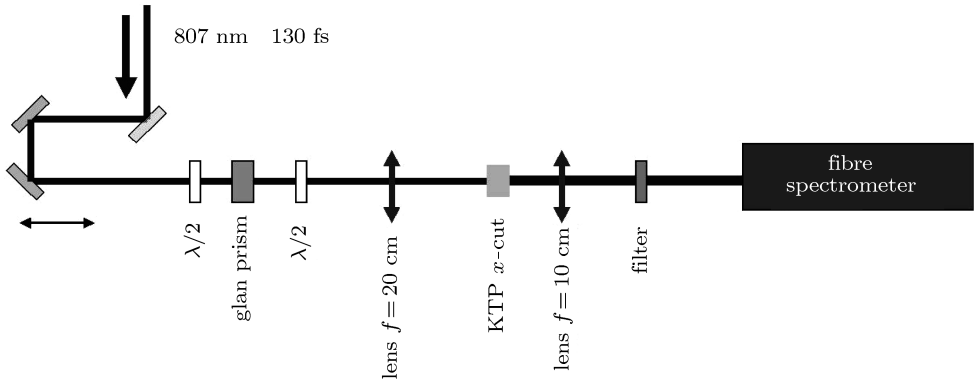


Fig. 1. Schematic diagram of experiment setup.

Figure 2(a) shows the spectrum of a 30 μJ laser pulse after passing through the KTP crystal with the polarization at an angle $\varphi = 45°$ with respect to the z (001) axis of the crystal. Filters were used in front of the spectrometer to attenuate the laser intensity in the 3.06 eV (404 nm) and 1.53 eV spectral regions. Besides SCG and SHG (3.06 eV), a new peak at 2.58 eV (480 nm) appears in the spectrum. The duration of the 2.58 eV pulse was measured by using a cross-correlator to be 500 fs, which is longer than its Fourier-transform limited pulse duration of 113 fs.

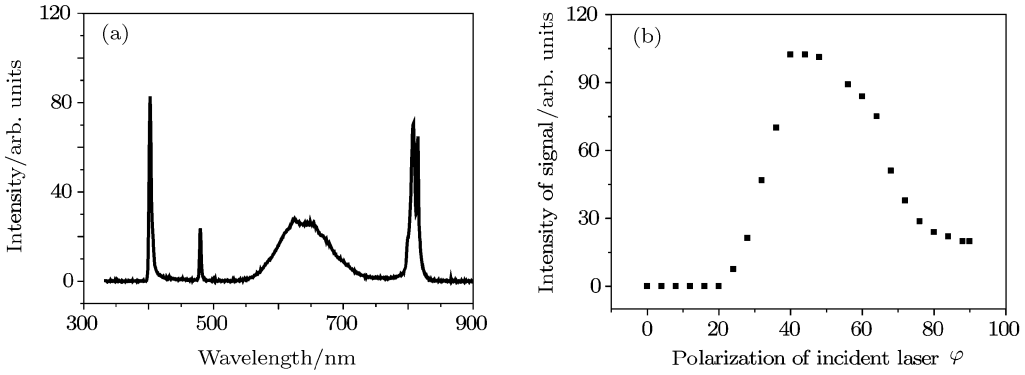


Fig. 2. Spectrum of the laser after passing through the crystal at $\varphi = 45°$, where φ is the angle between the polarization direction and the z axis of the crystal (a), and dependence of intensity of signal at 2.58 eV on polarization of the pump laser (b).

Figure 2(b) shows the intensity of newly generated signal at 2.58 eV at different values of φ. We can see that signal at 2.58 eV reaches a maximum when φ is about 45°. The polarization of the light at 2.58 eV and the SCG were detected to be along the y axis at any φ. The intensity dependence of the signal at 2.58 eV on incident laser energy is shown in Fig. 3. The intensity of 2.58 eV increases faster with a slope of 3.6 than that in Ref. [9] where the intensity of the sum frequency (SF) of the supercontinuum (SC) and the incident laser increased linearly with the intensity of the input pulse. The conversion efficiency was measured to be 3×10^{-5} when the pump energy was 49 μJ. It implied that a nonlinear polarization with an order larger than 3 might be involved.

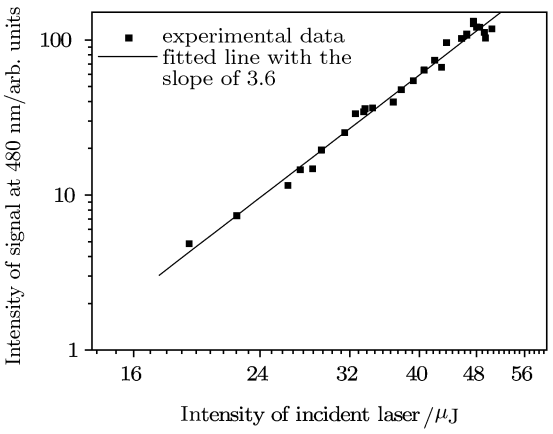


Fig. 3. Dependence of intensity of signal at 2.58 eV on the intensity of input laser pulse.

The results suggest that a wave mixing process

may be involved. To investigate this, we performed an amplification experiment with an injected seed laser beam as shown in Fig. 4. The fundamental laser beam is split into two beams, one serves as a pump beam and is polarized at $\varphi = 45°$, and the other is used to generate a super continuum (SC) in a water cell. The SC beam passes through two bandpass filters centred at 2.58 eV to ensure appropriate attenuation of the central pump beam spectral range and is focused together with the pump beam by a 20 cm achromatic focal length lens into the KTP crystal in a collinear configuration.

The 2.58 eV signal is amplified when the SC seed beam and the pump beam overlap spatially and temporally. The maximum amplification occurs when the seed beam is polarized along the y axis and no amplification is observed when the seed beam is polarized along the z axis. Figure 5 shows the amplified signal with using an incident pump energy of 19 μJ per pulse and an SC seed energy of 5 nJ per pulse. The spectra of the SC seed beam are also shown in Fig. 5. The resulting amplified signal energy at 2.58 eV is 20 nJ per pulse.

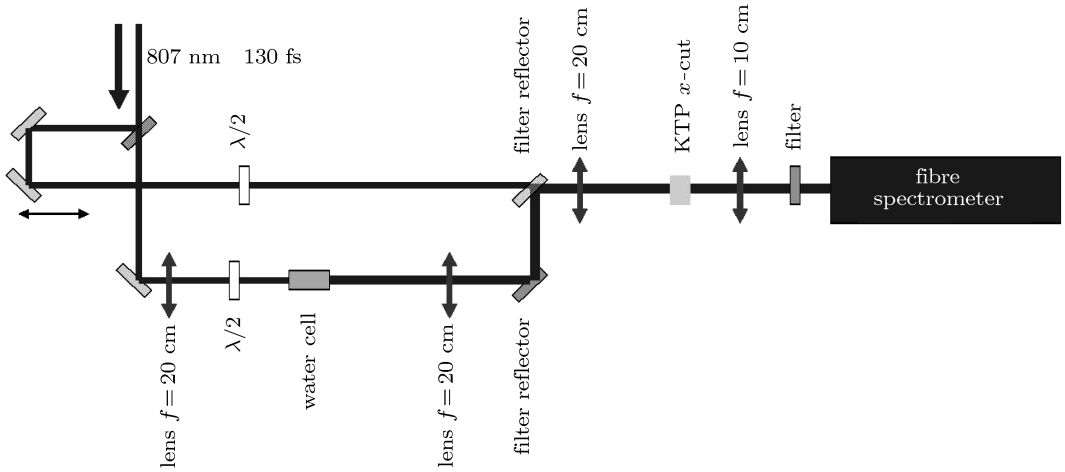


Fig. 4. Schematic diagram of experiment setup in an amplification configuration.

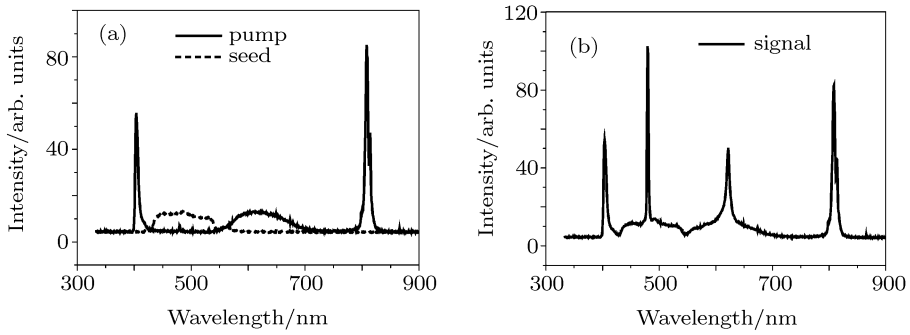


Fig. 5. Spectra of pump and seed beams containing 2.58 eV after passing through the crystal (a), and spectrum of amplified signal when pump and seed beams are overlapped spatially and temporally (b).

In addition to the 2.58 eV signal, a new amplified peak at 1.99 eV (622 nm) appears which is correlated with the 2.58 eV signal. To further investigate this, we change the filters in the seed beam path to inject into the KTP crystal an SC seed beam centred at 1.99 eV with an energy of 3 nJ per pulse. As shown in Fig. 6, the result shows an amplification at 2.58 eV and 1.99 eV each. These results suggest that we are observing a parametric amplification process where the signal is at 2.58 eV and the idler is at 1.99 eV.

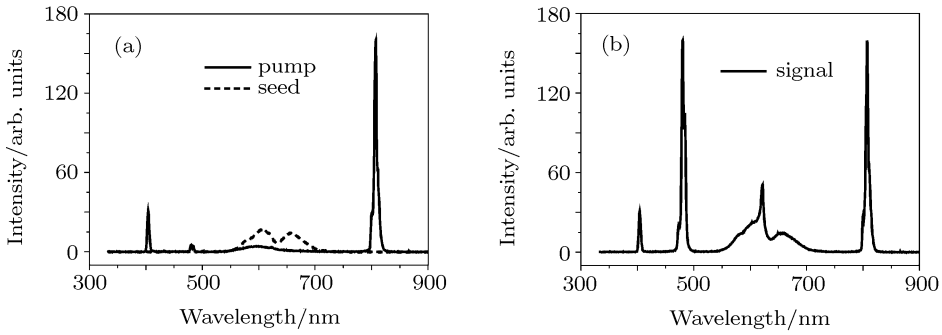


Fig. 6. Spectra of pump and seed beams containing 1.99 eV after passing through the crystal(a), and spectrum of amplified signal (b).

We hypothesize that the nonlinear optical process observed is based on the fourth-order nonlinear polarization

$$P_y^{(4)}(\omega_2) = \chi_{yyyzy}^{(4)}(\omega_2, \omega, \omega, \omega, -\omega_1) E_y(\omega) E_y(\omega) E_z(\omega) E_y^*(\omega_1), \tag{1}$$

in which ω corresponds to the fundamental pump beam frequency (1.52 eV), which is within the spectral range of the pump beam (centred at 1.53 eV), ω_1 to the signal frequency (2.58 eV), and ω_2 to the idler frequency (1.99 eV). This is a parametric frequency up-conversion process. The conservations of energy and momentum lead to

$$\omega_2 = \omega + \omega + \omega - \omega_1 \tag{2}$$

and

$$\boldsymbol{k}_y(\omega_2) + \boldsymbol{k}_y(\omega_1) = \boldsymbol{k}_z(\omega) + \boldsymbol{k}_y(\omega) + \boldsymbol{k}_y(\omega). \tag{3}$$

Equation 3 is satisfied within an error of 0.2% (we calculate the refractive index according to Ref. [14]).

The reader may note that Eq. (3) requires a pump beam polarized in both the y and the z directions, but in Fig. 2, the signal is not zero when $\varphi = 90°$. Hence we must explain how the signal at 2.58 eV is generated when the pump beam is apparently completely y-polarized. We suggest that z-polarized laser is generated by an energy transfer process from the y-polarized pump, or fundamental wave (FW), to a z-polarized FW, which occurs through SHG and OPO. Initially, a z-polarized second harmonic wave is generated by the y-polarized FW through the 2nd order process $P_z(2\omega) = \chi_{zyy} E_y^2(\omega)$, following which the z-polarized FW is generated by OPO through the second order process $P_z(\omega) = \chi_{zzz} E_z(2\omega) E_z^*(\omega)$. This energy transfer is possible due to the large value of χ_{zzz} and the small phase mismatch value in the OPO. This argument is supported by the fact that the z-polarized FW is detected after passing through the crystal to be at 4% with respect to the incident y-polarized FW.

We notice that, due to the focal position of the y-polarized pump beam within the KTP, it propagates a short distance in the crystal before SCG begins. When parametric amplification occurs, the y- and the z-polarized parts of the FW are separated in time because of the anisotropic refractive index of KTP, reducing the pump-to-signal conversion efficiency. In order to overcome this as shown in Fig. 7, we use polarization beams spplitters (PBS) to separate the two components with two different polarizations. Keep the y-polarized pump beam and inject a z-polarized FW pulse along the seed beam path. The time delay between the two pulses is adjusted to maximize the signal, and the experimental results are shown in Fig. 8.

When the FW y- and z-polarized pulse energies are 19 and 1 μJ, respectively, the signal pulse energy is 20 nJ, corresponding to a conversion efficiency of 0.1%. Thus the signal beam at 2.58 eV constitutes a new femtosecond pulsed laser source.

The intensity dependences of the signal on the y- and the z-polarized incident laser intensities are shown in Figs. 9(a) and 9(b) respectively. The curves are fit to power laws with slopes of 2.86 and 0.91, respectively, and the total intensity dependence coincides with that given in Fig. 3, indicating that the nonlinear process is the same in the two cases.

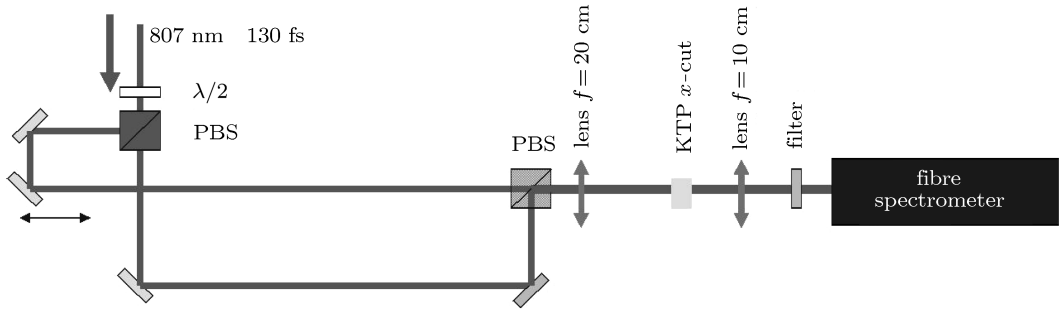


Fig. 7. Schematic diagram of experiment setup in an improving conversion efficiency configuration.

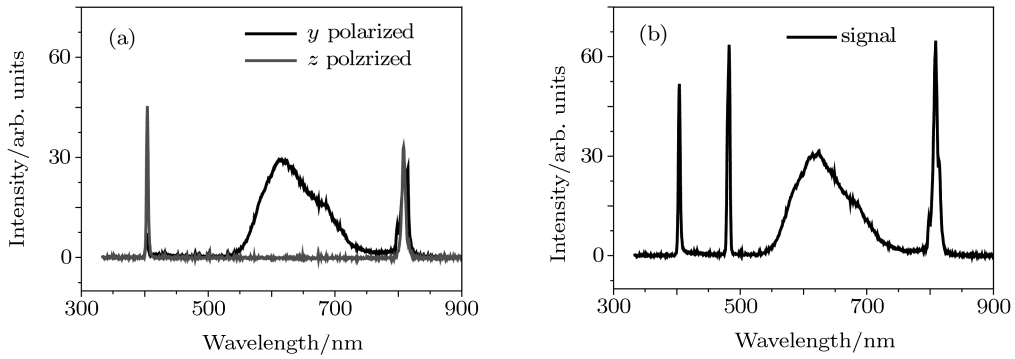


Fig. 8. Spectra of y-polarized and z-polarized lasers after passing through the crystal (a), and spectrum of amplified signal (b).

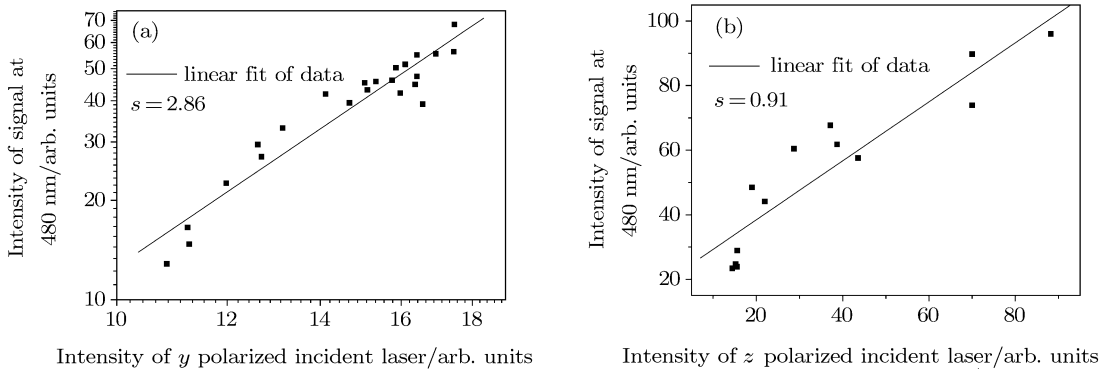


Fig. 9. Dependences of intensity of signal at 2.58 eV on the intensities of input y-polarized laser pulse (a) and z-polarized laser pulse (b).

As we know, the conversion efficient for the amplified signal depends on the intensity of incident laser and the thickness of crystal. The conversion efficiency can be improved by increasing the intensity of incident laser under the damage threshold of the crystal. For the KTP crystal, the y-polarized part will separate from the z-polarized part after it propagates 0.5 mm when one pulse with a duration of 100 fs is incident, a thicker crystal has nothing to do with the conversion efficiency. But in the case of two incident beams with different polarizations as shown in Fig. 7, lasers with different polarizations can overlap exactly, a thicker crystal will provide a better supercontinuum and give rise to the conversion efficiency.

We find that the intensity dependence of the signal on the 2.58 eV seed beam traverses a threshold when the energy per pulse of the seed beam reaches 6.8 nJ. Above this value as shown in Fig. 10, the energy per signal pulse decreases even as the energy per pulse of the SC increases. The simultaneous decrease of signal and idler implies a decrease in conversion efficiency. We suggest that the energy transfer from the amplified signal back to

the pump beam through the fourth-order polarization $P^{(4)}(\omega) = \chi^{(4)}(\omega, \omega_1, \omega_2, -\omega, -\omega)E(\omega_1)E(\omega_2)(E^*(\omega))^2$ is significant when $E(\omega_1)$ and $E(\omega_2)$ are large. This is a parametric frequency low-conversion process.

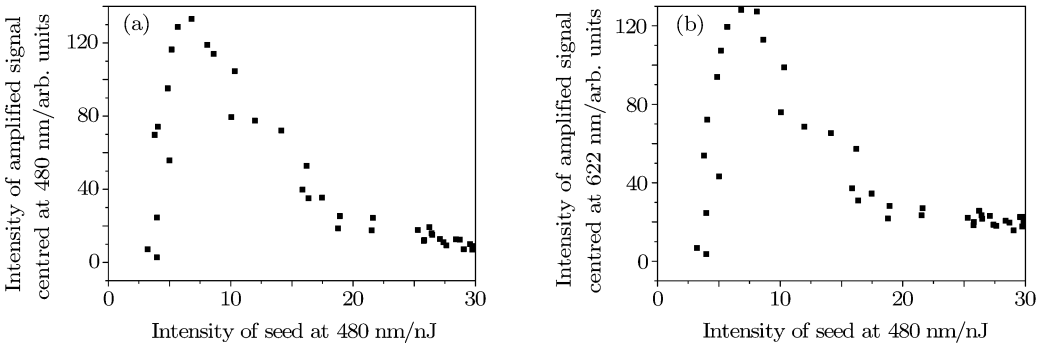


Fig. 10. Intensity dependences of amplified signal at (a) 2.58 eV and (b) 1.99 eV on the seed beam intensity centred at 2.58 eV.

In conclusion, we generate a new femtosecond pulsed source at 2.58 eV (480 nm) by parametric amplification in a KTP crystal with a fundamental wave at 1.53 eV (807 nm). SCG occurs through filamentation and supplies the optical seed pulses for the parametric amplification process. The conversion efficiency from pump to signal is 0.1% when we use two orthogonally polarized, collinear pump beams. When only y-polarized pump light is injected, z-polarized pump light is generated by cascade SHG and OPO process. If a seed contained signal is injected, both parametric frequency up-conversion and low-conversion processes through the fourth-order nonlinear polarization could be observed, depending on the seed beam intensity.

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