# Ultrabroadband noncollinear optical parametric amplification with LBO crystal

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**Abstract:** Ultrabroadband visible noncollinear optical parametric amplification (NOPA) was achieved in an LBO crystal, with a continuum seed pulse generated from a sapphire plate. The spectral bandwidth of the amplified visible pulse was about 200 nm, which can support sub-5 fs pulse amplification. An amplified output of 0.21 uJ with an average gain of about 210 was achieved. This provides, to the best of our knowledge, the first-time demonstration of such broadband amplification with a biaxial nonlinear optical crystal. Both the simulation and experimental results indicate that the LBO has a great potential as nonlinear medium in power amplifier for TW to PW noncollinear optical parametric chirped pulse amplification (NOPCPA) systems.



**OCIS codes:** (140.4480) Optical Amplifiers; (190.4410) Nonlinear Optics and Parametric Processes; (190.4970) Parametric Oscillators and Amplifiers

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## 1. Introduction

After the principle of optical parametric chirped pulse amplification (OPCPA) was first proposed by Dubietis *et al* [1], it has been an attractive tool to generate broadband laser pulses. OPCPA constitutes of an optical parametric amplification (OPA) pumped by a long pulse duration, used to amplify a large bandwidth and chirped pulse of nearly equal pulse duration compared with pump pulse width. This amplification scheme has several advantages, such as high gain with broad bandwidth, high signal-noise contrast ratio and small B integral. In a broadband OPA system, the group velocity of signal and idler pulse must be matched. There are two types of geometries to realize a broadband OPA; collinear and noncollinear geometries. For collinear geometry, a broadband type-I OPA can only be realized at degeneracy, because the group velocity of signal and idler pulses can be matched only at the degeneracy; usually uniaxial or biaxial nonlinear optical crystals are used, such as LBO [2], BIBO [3], BBO [4], CLBO [5], and KDP [6] crystals. A 1um OPCPA system with 16.7 TW/120 fs output using LBO crystals was demonstrated by Xu *et al* [7]. For noncollinear geometry, a broadband OPA is realized at the nondegeneracy, the noncollinear angle of pump and signal must meet the group-velocity matching condition between signal and idler, which is equivalent to an achromatic phase matching with the spectral angular dispersion of the idler. It usually uses uniaxial optical crystals: KD*P and BBO [8]. Lozhkarev *et al* [9] used KD*P crystal to setup an OPCPA system with 200 TW/45 fs. Baltuska *et al* [10] obtained a 3.9 fs visible pulse from a NOPA based on a BBO crystal. Witte *et al* [11] generated a 2 TW/7.6 fs laser pulse using NOPA, with a BBO crystal. An LBO crystal was used by Liu *et al* [12] to generate broad amplification with NOPCPA, and the amplified bandwidth was about 53 nm.

In this article, we reported the experimental results for the generation of an ultrabroadband visible laser pulse from a NOPA based on an LBO crystal, pumped with ~350 fs, centered on 392.5 nm and possessing a 3 nm (FWHM) spectral bandwidth. The bandwidth of the amplified pulse was about 200 nm (FWHM). Furthermore simulation shows that LBO crystal can support sub-5 fs pulse amplification with NOPCPA, pumped by 526.5 nm. Therefore LBO has a great potential to be used as energy amplifier for TW or PW of NOPCPA with sub-5 fs pulse.

## 2. Theoretical analyses and discussion

Firstly, the theoretical equations are deduced. Then the NOPA gain spectra of LBO with pump wavelength around 392.5 nm are simulated in order to compare with experimental results. At last, the NOPA gain spectra of LBO and KD*P are simulated with pump wavelength of 526.5 nm and compared together.

Since OPA is a typical three-wave coupled nonlinear process, conservation of energy and momentum is required to be satisfied as given by the following equations,

#98774 - $15.00 USD Received 14 Jul 2008; revised 24 Aug 2008; accepted 17 Sep 2008; published 31 Oct 2008

$$\hbar w_p = \hbar w_s + \hbar w_i,$$
$$\vec{k}_p = \vec{k}_s + \vec{k}_i, \quad (1)$$

where the subscripts p, s and i refer to pump, signal and idler lights, respectively.

Here we restrict our attention to noncollinear interactions in xy plane of LBO crystal, in the principal-axis coordinate, as shown in Fig. 1. The three-wave interactions satisfy the type I ($e_1$+$e_1$→$e_2$) phase matching condition in an xy plane ($e_1$ means a slow light and $e_2$ means a fast light). Pump wave is polarized and propagates in the xy plane, and signal and idler are both polarized along the z axis. Signal has a small non-collinear angle $\alpha$ with pump light, and pump vector has a phase-matching angle $\theta$ with x axis.

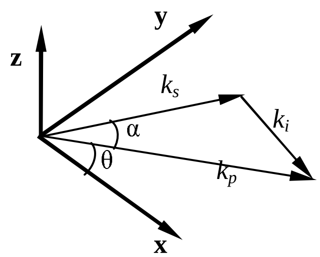


Fig. 1.Geometry of noncollinear phase matching in xy plane.

From Eq. (1), the fundamental expressions for noncollinear three-wave mixing are derived as,

$$1/\lambda_p = 1/\lambda_s + 1/\lambda_i,$$
$$\left[\frac{n_i}{\lambda_i}\right]^2 = \left[\frac{n_p}{\lambda_p}\right]^2 + \left[\frac{n_s}{\lambda_s}\right]^2 - 2\cos\alpha \frac{n_s n_p}{\lambda_s \lambda_p}, \quad (2)$$
$$\frac{1}{n^2{}_p} = \frac{\cos^2\theta}{n^2{}_{py}} + \frac{\sin^2\theta}{n^2{}_{px}},$$

Here $n_{px}$ and $n_{py}$ are the principal refractive indexes of x and y axis at pump wavelength, respectively.

For a uniaxial KD*P crystal, the fundamental expressions are the same as those in Eq. (2), just $n_{px}$ and $n_{py}$ are replaced with $n_{po}$ and $n_{pe}$, respectively. Here, $n_{pe}$ and $n_{po}$ are the principal extra-ordinary and ordinary crystalline indexes of refractive for pump wavelength, respectively. $\theta$ is the phase-matching angle of the pump relative to the optical axis.

The phase mismatching $\Delta k$ can be defined as [13],

$$\Delta k(\lambda_s) = \left|\Delta\vec{k}(\lambda_s)\right| = 2\pi\left(\frac{n_i(\lambda_s)}{\lambda_i(\lambda_s)} - \frac{n_{io}(\lambda_{so})}{\lambda_{io}(\lambda_{so})}\right), \quad (3)$$

where $\lambda_{io}$ and $\lambda_{so}$ are the ideal phase matched idler and signal wavelength, respectively.

The intensity gain of the amplified signal beam can be obtained using the analytical solution of the coupled wave equations in the slowly varying envelope approximation, assuming no significant pump depletion and the group-velocity mismatching (GVM) neglected. The intensity gain (G) is given [14] by

$$G = 1 + (\xi L)^2 (\sinh B / B)^2, \quad (4)$$

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where $\xi\left(=4\pi d_{eff}\sqrt{I_p/2\varepsilon_0 n_p n_s n_i c\lambda_s\lambda_i}\right)$ is the effective gain coefficient, $B=\sqrt{(\xi L)^2-\left(\Delta kL/2\right)^2}$, and $L$ is the crystal length, $d_{eff}$ is the effective nonlinear coefficient, and $I_p$ is the pump intensity.

Based on the above equations and the Sellmeier equations of LBO [15] and KD*P (with 96% deuteration level) [16], the gain spectra can be calculated. The NOPA gain spectra of 1 mm thick LBO in noncollinear geometry were calculated as shown in Fig. 2, with pump wavelength of about 392.5 nm; the pump intensity is 200 GW/cm$^2$. Fig. 2 shows that the maximal gain bandwidth of LBO is about 220nm (FWHM) with pump at 392.5 nm, the corresponding noncollinear angle and phase matching angle is 2.84$^o$ and 37.58$^o$, respectively. Under the same condition ($\alpha$=2.84$^o$, $\theta$=37.58$^o$), the gain spectra are also shown in Fig. 2, with pump 391 nm and 394 nm.

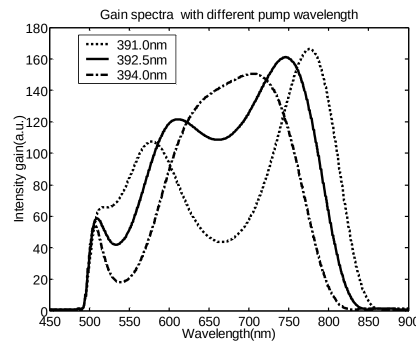


Fig. 2. Gain spectra of NOPA in xy plane with three different pump wavelengths of 391 nm, 392.5 nm, and 394 nm; with $\alpha$=2.84$^o$ and $\theta$=37.58$^o$.

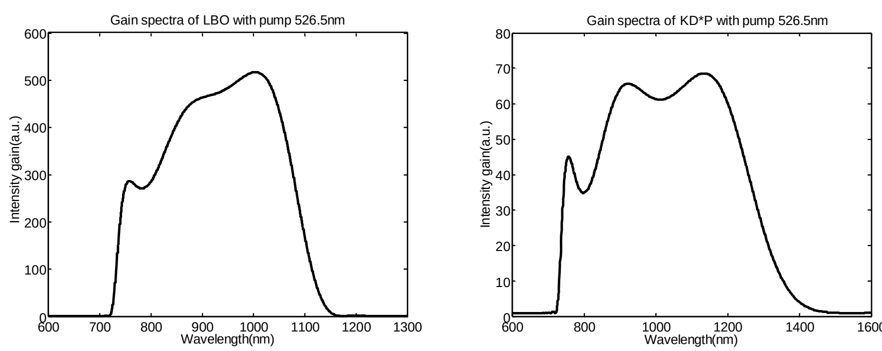


Fig. 3. Optimal NOPA gain spectra of LBO and KD*P with pump 526.5 nm.

The gain spectra for LBO and KD*P in nocollinear geometry are also calculated as shown in Fig. 3, with pump wavelength of 526.5 nm, 5 mm long LBO & KD*P, and the pump intensity is 10 GW/cm$^2$ and 80 GW/cm$^2$, respectively. The optimal gain bandwidth for LBO and KD*P are about 337 nm and 530 nm, and the corresponding noncollinear angles are 1.31$^o$ and 0.54$^o$, respectively. The simulated results show that LBO & KD*P can support sub-5 fs pulse amplification. Because the effective nonlinear coefficient of LBO is more than 2 times larger than that of KDP and KD*P crystals [15], the intensity gain of LBO is much higher than that of KD*P under similar operating conditions. Recent development of crystal growth technology has enabled LBO crystal to be grown up to the size of 146×145×62 mm$^3$ [17], and

#98774 - $15.00 USD Received 14 Jul 2008; revised 24 Aug 2008; accepted 17 Sep 2008; published 31 Oct 2008

also its damage threshold is higher than that of KDP and KD*P crystal [18], therefore LBO is more suitable to be used as energy amplifier for TW to PW of NOPCPA with sub-5 fs pulse.

### 3. Experiment

Our NOPA experimental setup is shown in Fig. 4. A Bright CPA system was used as a source, delivering 130 fs pulses with 1.35 mJ energy at 785 nm and 1 kHz repetition rate. For the continuum seed of the NOPA, a fraction of 27 uJ of 785 nm pulse was split off and its intensity was controlled with a variable neutral-density filter (VND); then the polarization was converted to vertical polarization using a half-wave plate. The appropriate energy was focused with an *f*=100mm lens to a 2 mm thick sapphire to generate single-filament femtosecond continuum seed pulse. The continuum seed pulse was precompressed to ~200 fs by a pair of parallel gratings, introducing negative dispersion of about 350 fs between 500 nm and 800 nm. The distance between two parallel gratings with 150 *l*/mm was 12 mm. The long wavelength part of continuum seed pulse was cut off through a 1 mm filter. The spectrum after filter was used as seed and shown in Fig. 5. For the pump of NOPA, about 200 uJ of 785 nm pulse was used to generate SHG through a 0.4 mm thick BBO crystal in type-I configuration. The SHG was stretched to about 350 fs by a 150 mm-long fused silica block. The polarization of SHG was changed to a horizontal polarization using a half-wave plate. The bandwidth (FWHM) and energy of SHG was 3 nm and 45 uJ energy, respectively.

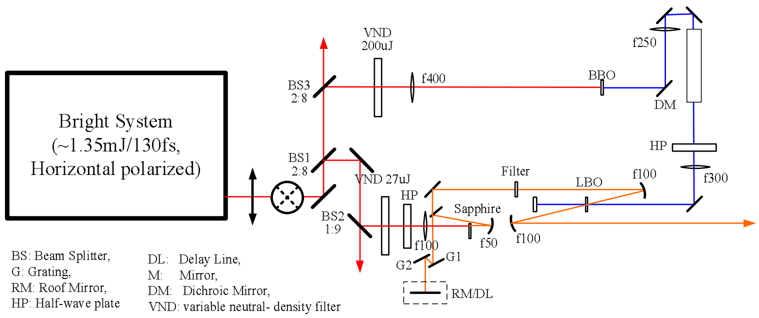


Fig. 4. Layout of NOPA experiment

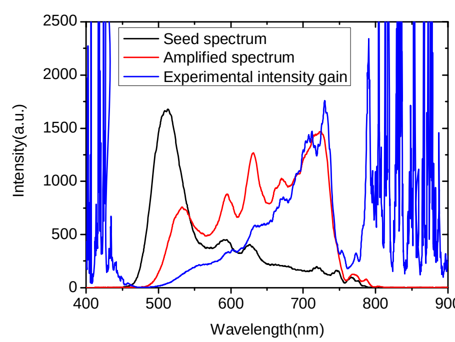


Fig. 5. The spectrum of continuum seed (black line) before NOPA; The spectrum of amplified spectrum (red line) after NOPA; The experimental gain spectrum (blue line).

The femtosecond continuum seed was then noncollinearly amplified at the noncollinear angle $\alpha$=2.84$^o$ inside the LBO crystal with cutting angle of $\theta$=37$^o$, for the signal-idler group-velocity matching. The pump and signal pulses were aligned and focused for maximum overlap over the whole crystal length, with *f*=300 mm lens and *f*=100 mm concave mirror, respectively. The energies of signal and pump were 1 nJ and 40 uJ, respectively. In order to

#98774 - $15.00 USD Received 14 Jul 2008; revised 24 Aug 2008; accepted 17 Sep 2008; published 31 Oct 2008

obtain maximum bandwidth and energy of amplified pulse, the delay between signal and pump, the noncollinear angle and crystal angle were all finely adjusted. When the spectrum bandwidth of amplified signal pulse reached maximum, the amplified signal pulse energy was about 0.21 uJ, the energy fluctuation was below 5%. The average gain G was about 210. The signal pulse spectra before and after amplification were shown in Fig. 5. It shows that the amplified signal has a spectral bandwidth (FWHM) of about 200 nm, except a dip at 575 nm. To our knowledge, such ultrabroadband amplified spectrum using biaxial crystal is firstly reported.

The experimental intensity gain spectrum is also shown in Fig. 5, by dividing amplified spectrum to seed spectrum. It shows noisy errors existing below ~450nm and above ~785nm, because of low measured spectral intensity of seed and amplified pulses. The intensity gain exists from ~500nm to ~770nm, although the bandwidth of experimental intensity gain (FWHM) is 60nm. The main reasons for such experimental intensity gain are that the stretched pump pulse is Gaussian temporal profile; the maximal amplified spectrum is obtained by changing delay between signal and pump pulse; and there exists high order dispersion in seed pulse. Because the full width of experimental intensity gain spectrum is broader than 200nm, it can be concluded that an LBO is suitable for sub-5 fs NOPA system.

Comparison between Figures 2 and 5 shows that the experimental intensity spectrum is basically consistent with that calculated for a pump wavelength of 392.5 nm. Therefore we can conclude that the spectral bandwidth of the pump has no substantial effect on the gain spectra of the NOPA. Because the pump pulse width (FWHM) is about 350 fs, signal pulse width is about 200 fs with negative dispersion, the GVMs between pump (392.5 nm) and signals at 500 nm and 800 nm were calculated to be 25 fs and 128 fs in a 1 mm thick LBO crystal, therefore the GVM can be neglected in the calculation with Eq. (4), to obtain the results shown in Fig. 2.

## 4. Conclusions

In conclusion, we developed a NOPA system based on a LBO crystal, the average gain G of LBO, with ultra broadband of 200 nm, was about 210. Furthermore, we simulated the intensity gain of NOPA pumped by 526.5 nm. The gain bandwidth is about 337 nm, it can support sub-5 fs pulse amplification. Now LBO crystal with size larger than 100 mm can be grown. It has large damage threshold and effective nonlinear coefficient, compared with KDP and KD*P. Therefore it has a great potential as the energy amplifier for TW to PW of sub-5 fs NOPCPA.

## Acknowledgments

This work was partly supported by the grant from the Ministry of Education (MOE) in Taiwan under the ATU Program at National Chiao Tung University. A part of this work was performed under the joint research project of the Laser Engineering, Osaka University under contract subject B 1-27.

#98774 - $15.00 USD Received 14 Jul 2008; revised 24 Aug 2008; accepted 17 Sep 2008; published 31 Oct 2008