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Research Article

A Novel Femtosecond Laser System for Attosecond Pulse Generation

Jianqiang Zhu, Xinglong Xie, Meizhi Sun, Qunyu Bi, and Jun Kang

National Laboratory on High Power Laser and Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Number 390, Qinghe Road, Jiading, Shanghai 201800, China

Correspondence should be addressed to Xinglong Xie, xiexl329@mail.shcnc.ac.cn

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We report a novel ultrabroadband high-energy femtosecond laser to be built in our laboratory. A 7-femtosecond pulse is firstly stretched by an eight-pass offner stretcher with a chirp rate 15 ps/nm, and then energy-amplified by a two-stage optical parametric chirped pulse amplification (OPCPA). The first stage as preamplification with three pieces of BBO crystals provides the majority of the energy gain. At the second stage, a YCOB crystal with the aperture of ~50 mm is used instead of the KDP crystal as the gain medium to ensure the shortest pulse. After the completion, the laser will deliver about 8 J with pulse duration of about 10 femtoseconds, which should be beneficial to the attosecond pulse generation and other ultrafast experiments.

The attosecond pulse has been of interest in recent years and proved to be a powerful tool in studying the ultrafast phenomena, such as the chemical/biological transformations occurring on the femtosecond timescale and the evolution of the dynamics of the electrons in atoms or molecules irradiated by intense lasers. There are two ways to generate an attosecond pulse/pulse train. One is through the nonlinear processes of the superposition of high order harmonics generated in the laser-gas atom interactions in which the laser intensity should be low so as to avoid the ionization of atoms. Up to now, the laser intensity is no more than 10¹⁶ W cm⁻² and the efficiency of harmonic emission from the atoms is low. The other way is to generate the high order harmonics from the dense surface of a plasma created by the high-intensity femtosecond laser, which is promising for the intense attosecond pulse generation. Recent experiments and simulations have shown that attosecond pulses with high conversion efficiency, high photon energy, and excellent divergence can be generated [1–4].

The efficiency of harmonic generation from the laser-plasma surface relies on the laser intensity. In the experiments for the brilliant harmonic generation from the surface, the laser has energy of 0.1 J to less than 10 J with a pulse duration of 30–50 fs [5]. In this paper, we report a novel

ultrabroadband ultrashort high-energy laser system based on optical parametric chirped pulse amplification (OPCPA). After completion, the laser can deliver 8.0 J with pulse width of about 10 fs, which is much shorter than those used in the previous intense attosecond pulse generation experiments. The laser should be beneficial to the attosecond pulse generation and other ultrafast experiments.

OPCPA has been applied and becomes promising for constructing high-intensity lasers [5-9], particularly with the invention of new nonlinear crystals such as YCOB (YCa₄O(BO₃)₃, YCOB) [10-12]. Compared with other nonlinear crystals commonly utilized in OPCPA systems, YCOB crystal has several prominent properties: high damage threshold, moderate thermal conductivity, and nonlinear coupling [13]. It has been shown that the size of YCOB crystal can be as large as 7.5 cm in diameter and 25 cm in length [14], much larger than those currently used, such as LBO and BBO crystals. Chekhlov et al. reported their OPCPA laser system which used large aperture KDP crystals for power amplification. Their laser delivers output energy up to 35 J and pulse width of 84 fs, [15]. To our knowledge, YCOB crystal has not been used in OPCPA laser systems with wavelength centered at 808 nm and output energy up to several joules and pulse width of less than 10 fs.

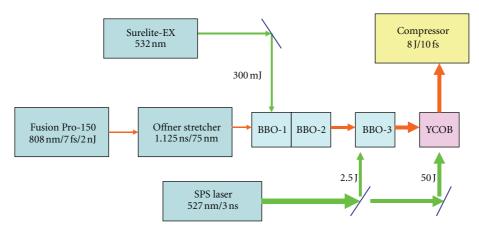


FIGURE 1: Optical schematic layout of OPCPA.

To design the OPCPA-based ultrashort laser system which can deliver several joules within about 10 fs, we use YCOB instead of KDP or DKDP crystal as the gain medium for power amplification stage. Because YCOB has nonlinear coefficient four times of that of KDP and DKDP, it can provide broad spectral gain bandwidth. As indicated in Figure 1, our laser system consists of five parts: a signal laser pulse, an Offner stretcher, a multistage optical parametric amplifier (OPA), a compressor, and a pump laser system. The oscillator provides signal pulses of 7 fs in time duration and 2 nJ in energy. The signal pulses are chirped to 15 ps/nm by an eight-pass Offner stretcher with 10 percent transmission efficiency, and the energy of chirped signal pulse injected into OPA chain is measured to be about 0.2 nJ. The OPCPA amplification chain is composed of two stages. The first stage, as a preamplification, includes three pieces of BBO crystals that provide the majority of the energy gain. The pump pulses at 532 nm with 300 mJ for the first two pieces of BBO crystals are from a commercial single longitudinal mode Nd:YAG pump laser which operated at a repetition of 10 Hz. For the third piece of BBO crystal, 2.5 J second harmonic pulse, from an Nd:glass Subpicosecond Laser System (SPS), is utilized as the pump source [16]. The SPS laser is a two beam system, one beam is short with pulse width ~1.0 ps and pulse energy 100 J. The other beam is a 1053 nm beam with the pulse width of about 2.0 ns and maximum pulse energy about 150 J. The nanosecond beam is used as the pump source for our OPCPA system. The whole system works at one shot every thirty minutes. In the second stage, a piece of YCOB crystal with large aperture ~50 mm is utilized as the gain medium and the SPS provides pulses at 526.5 nm with the desired special-temporal shape and energy exceeding 50 J.

The signal is amplified in the XOZ principle of YCOB because the effective nonlinear coefficient (d_{eff}) in XOZ principle plane is much larger than that in XOY and YOZ principle planes [17, 18].

Noncollinear geometric configuration with type 1 phase matching $o + o \rightarrow e$ is utilized in all OPA stages. Compared with the collinear geometric configuration, the noncollinear geometric configuration can achieve larger parametric bandwidth and get higher conversion efficiency by compensating

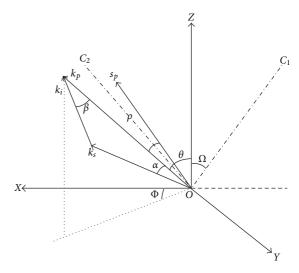


FIGURE 2: Noncollinear phase matching vectors in biaxial crystals.

walk-off effects of the pump pulse with noncollinear angle. Wave vectors and parameters of noncollinear phase matching in biaxial crystals are shown in Figure 2. X, Y, and Z are principle axes. The refractive indexes along principle axes are ordered nZ > nY > nX. C_1 and C_2 are optical axes. Ω is optic axial angle, Φ the azimuth of k_p in XOZ principle plane, and k_s , k_i , and k_p wave vectors of signal, idle, and pump, respectively. Symbol of α refers to the noncollinear angle, θ the phase matching angle, ρ the walk-off angle, s_p the Poynting vector of pump pulse, and θ the angle between k_i and k_p . Momentum conservation and energy conservation during the OPA process can be expressed as (1) and (2). The wave vector mismatch is expressed as (3):

$$\overrightarrow{k}_p = \overrightarrow{k}_i + \overrightarrow{k}_s, \tag{1}$$

$$\omega_p = \omega_i + \omega_s, \tag{2}$$

$$\Delta k = k_p - k_s \cos \alpha - k_i \cos \beta. \tag{3}$$

The optimized noncollinear configuration can realize the highest parametric bandwidth $(\Delta \lambda_p)$ for the ultrabroadband optical parametric amplifier. Generally speaking,

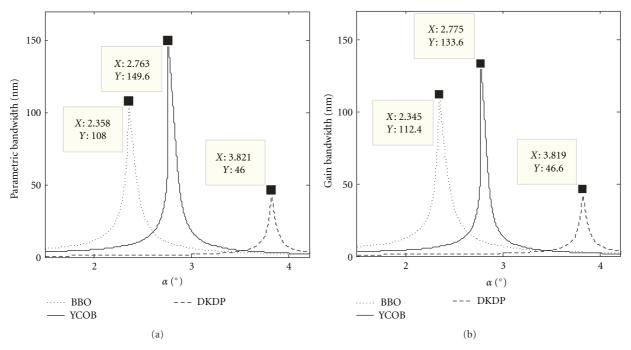


FIGURE 3: Parametric bandwidth (a) and gain bandwidth (b) versus noncollinear angles in BBO (dotted), in the XOZ principle plane of YCOB (solid), and in DKDP (dashed).

noncollinear angle in such configuration is set to realize group velocity matching (GVM) between signal pulse and pump pulse [19]. However, according to numerical analysis, there exists a small difference between the GVM noncollinear angle and the one for realizing the highest parametric bandwidth [20]. Parametric bandwidths as well as gain bandwidths ($\Delta \lambda_g$) and acceptance angles ($\Delta \theta$) are calculated exactly according to the definitions without using approximation of Taylor series. Identical analytic method was utilized in reference [18].

Parametric bandwidth is defined as the interval of the signal wavelength that restricts the phase mismatching smaller than $\pm \pi$ under the condition that perfect phase matching is achieved at the center wavelength of OPA. It can be expressed as $|\Delta kL| \leq \pi$, here L is the crystal length. Figure 3(a) shows the curves of the parametric bandwidths versus noncollinear angles in 15 mm BBO crystal, 15 mm YCOB crystal, and 35 mm DKDP. As shown in Figure 3(a), parametric bandwidths of 149.6 nm and 108 nm are achieved in YCOB and BBO when noncollinear angle are 2.763° and 2.358°, respectively. The value is 46 nm in DKDP when noncollinear angle is 3.821°. Parametric bandwidth is consistent with the gain bandwidth to a large extent. Gain bandwidth is defined as the interval of the signal wavelength that restricts the gain bigger than half of that achieved at the center wavelength of signal pulse. As shown in Figure 3, the largest gain bandwidth of 133.6 nm is achieved at noncollinear angle of 2.775° in YCOB, 112.4 nm at 2.345° in BBO, and 46.6 nm at 3.819° in DKDP. As a conclusion, gain bandwidths of YCOB and BBO are broad enough for the amplification of chirped pulses, while DKDP is confided to the amplification of chirped pulses with original pulse duration exceeding 21 fs at 808 nm.

The walk-off effect results from the separation of Poynting vector and wave vector of light transmitting in nonlinear crystals (inhomogeneous medium) and leads to spatial displacement between the signal pulse and pump pulse and the degradation of conversion efficiency in OPCPA. In addition, walk-off effect plays an important role in the impact of wavefront phase distortion of pump on the beam quality of signal in OPA [21, 22]. The acceptance angle is defined as the interval of θ that restricts the wave vector mismatch smaller than $\pm \pi/L$ around the perfect phase matching angle (θ_{pm}) at 808 nm. A large acceptance angle means a stable OPA. The walk-off angles, acceptance angles, and phase-matching angles versus noncollinear angles are shown in Figure 4. Corresponding to the optimal noncollinear angle, the walkoff angle in YCOB crystal is 1.134°, as well as acceptance angle ~0.06° and phase matching-angle 26.4°. The values of YCOB, BBO, and DKDP are listed in Table 1 and that the YCOB has bigger acceptance angle and smaller walk-off angle than BBO and DKDP.

As shown in Figure 5(a), for chirped signal pulse pumped by 532 nm pulse at intensity (I_p) of $0.4\,\mathrm{GW/cm^2}$ in 15 mm BBO crystal, gain of 2732 can be realized. In the first two pieces of BBO crystals, unsaturated gain of 1000×1000 can be achieved by pumping with 532 nm pulse of 300 mJ at an intensity of $0.4\,\mathrm{GW/cm^2}$. In the third piece of BBO crystal, the injected signal pulse is amplified by 526.5 pump pulse with energy of 2.5 J at an intensity of $0.4\,\mathrm{GW/cm^2}$. The phase matching parameters listed in Table 1 are calculated with the same method. There are not big differences with those pumped by pulse at 532 nm. The BBO crystal utilized in this stage is 13 mm (aperture) \times 15 mm (length) to realize saturated gain of \sim 2000 so that energy of signal pulse is amplified up to \sim 0.4 J. The majority of energy is provided

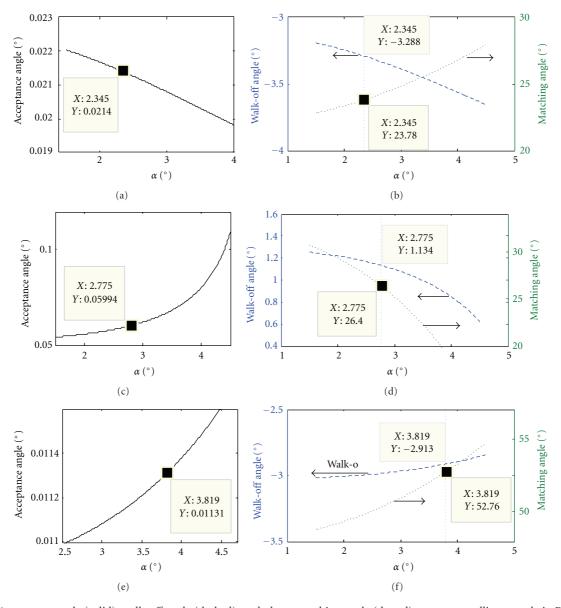


FIGURE 4: Acceptance angle (solid), walk-off angle (dashed), and phase-matching angle (dotted) versus noncollinear angle in BBO (a and b), the XOZ principle plane of YCOB (c and d), and DKDP (e and f).

Table 1: Noncollinear phase matching parameters of BBO, YCOB, and DKDP.

	α (°)	$ heta_{ m pm}$ (°)	$\Delta \lambda_p \text{ (nm)}$	$d_{\rm eff}$ (pm/V)	ρ (°)	$\Delta heta$ (°)	$\Delta \lambda_g (nm)$	I_p (GW/cm ²)	L(m)
BBO-1/2	2.345	23.78	108	2.096	-3.288	0.0214	112.4	0.4	0.015
BBO-3	2.4	23.86	132	2.095	-3.295	0.0214	127	0.4	0.015
YCOB	2.775	(26.4, 180)	149.6	0.935	1.134	0.0599	133.6	0.5	0.015
DKDP	3.819	52.76	46	0.295	-2.913	0.0113	46.6	2	0.035

by the second stage. Figure 5(b) shows the gain curve in the XOZ principle plane of YCOB with 526.5 nm pump pulse. For YCOB crystal of 15 mm in length, saturated gain of ~20 can be realized with pump pulse at intensity of 0.5 GW/cm² and energy of 50 J. In the power amplification stage, signal pulse of ~10 J can be achieved with broad spectral bandwidth. Moreover, saturated amplification in

both stages would greatly improve the stability of the system [23]. Normalized gain curve in 35 mm DKDP is also shown in Figure 5(a) when pumped by 526.5 nm pulse at intensity of 2 GW/cm². DKDP is a good choice for amplifying pulses centered at 808 nm with duration exceeding 20 fs.

As the temporal profile is an important factor in the ultrashort laser mater interaction experiment, a spectrum

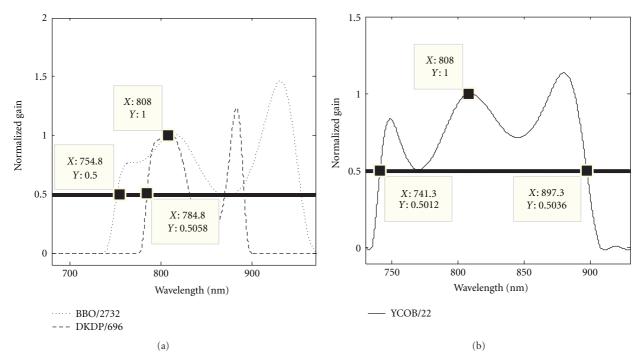


FIGURE 5: Normalized gain curves in BBO (a) (dotted), DKDP (a) (dashed), and XOZ principle plane of YCOB crystal (b) (solid).

shaper or a Dazzler with the resolution of 0.6 nm is used within the first OPCPA stage to control the pulse envelop. A 39-element deformable mirror with diameter of 50 mm is used before the pulse compression to ensure a good beam quality and the shortest pulse width.

The compressor is consisted of a pair of gratings and a piece of reflector. They are arranged in tandem in a four-pass configuration with Littrow incident angle of 36.3° to match with the stretcher so that the chirped pulse can be compressed to Fourier transform limitation. The transmission efficiency of the compressor is ~75% and the energy of output ultrashort pulse would be ~8 J.

In conclusion, a new type of OPCPA system to realize the amplification of pulse centered at 808 nm is analyzed, and noncollinear phase matching parameters of YCOB is compared with those of BBO and DKDP in Table 1. Numerical analysis shows that YCOB rather than DKDP can be utilized to amplify ultrabroadband chirped pulse so that the signal pulse can be compressed to about 10 fs after the compressor. With YCOB as the gain medium of OPCPA, a total gain of $\sim 4 \times 10^{10}$ is achieved and the energy of the amplified signal pulse is up to several joules.

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