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Generation of tunable ultrafast ultraviolet third harmonic by collinear compensation of group-velocity mismatch

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A B S T R A C T

We demonstrate a high efficient frequency tripling configuration of Ti: sapphire amplifier system for wavelength-tunable ultrafast ultraviolet laser generation. A new nonlinear crystal $Ba_{1-x}B_{2-y-z}O_4Si_xAl_yGa_z$ and a type-II phase-matched β-BaB_2O_4 crystal are employed for the second and the third harmonic generation, respectively. Significant improvement in conversion efficiency of frequency tripling is achieved by using a 65°-cut, 3-mm-long β-BaB_2O_4 crystal as the collinear group velocity compensation plate. Tunable ultraviolet pulse within the wavelength range from 256.7 to 276.7 nm have been produced, with a maximum average power of 212 mW, corresponding to a conversion efficiency of 8.48% for the third harmonic generation with 2.5 W fundamental power. The maximum pulse energy of the third harmonic is up to 0.21 mJ and it is estimated that the peak power is above 1 GW at 266.7 nm.

1. Introduction

Tunable coherent ultraviolet (UV) light sources, especially ultrashort pulse UV lasers are of great applications in the fields of material processing, time-resolved spectroscopy, laser spectroscopy and laser fusion. At present, ultrashort pulses are produced generally from passively mode-locked laser based on solid-state or fiber gain medium, such as Ti:Sapphire, ytterbium-doped yttrium aluminum garnet (Yb:YAG) and Yb-doped fiber. With these approaches, however, the wavelength of the output pulse is limited by the gain mediums, lying in the visible and near-infrared spectra regions. In the UV region, the best choice for the generation of the laser pulses in the femtosecond or picosecond regime is the frequency up-conversion technique based on second order nonlinearities [1–3], such as the second harmonic (SH) generation, the third harmonic (TH) generation and the fourth harmonic (FH) generation. For the TH generation process, the most widely used method is collinear sum-frequency mixing (SFM) configuration, which works well for continuous wave or relative long laser pulse. However, this approach is not suitable for the TH generation in the femtosecond regime due to the group-velocity mismatch (GVM) between the interacting waves. The GVM will decrease the conversion efficiency and broaden the pulse duration since the SFM only occurs when the interacting pulses overlap each other. One suggested method to compensate the GVM is to adjust the path length between the fundamental and SH pulses by means of time delay line consisting of beam splitters and mirrors [4]. This method has proven effective for some application, but is very difficult to align, and need precise translation stage to adjust the time delay. Another approach is to introduce one or multi plates with anomalous dispersion to compensate the group velocity walk-off [5–10]. In 2009, Wang et al. [11] demonstrated a collinear configuration of doubling-compensation-tripling system for high efficiency of TH generation. A 29.2°-cut, 0.2-mm-long BBO crystal which provided anomalous dispersion is used as compensation plate to compensate group velocity delay. 1.6 times improvement of conversion efficiency for the TH generation is achieved comparison with conventional configuration.

Over the past three decades, a lot of nonlinear crystals, such as LiB_3O_5 (LBO), β-BaB_2O_4 (BBO), $CsLiB_6O_{10}$ (CLBO) [12], $KBe_2BO_3F_2$ (KBBF) [13–15] and $Ba_{1-x}B_{2-y-z}O_4Si_xAl_yGa_z$ (BBSAG) [16], have been developed and employed for the generation of femtosecond pulses in the blue, UV, and even the deep-ultraviolet (DUV) region through frequency up-conversion technique. Among these crystals, BBO is considered as an excellent nonlinear crystal for the UV and DUV femtosecond pulses generation due to its high nonlinear coefficient, high optical damage threshold, and broad transparent

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range extends down to 189 nm [17]. BBO also exhibits anomalous dispersion effect cut with a suitable angle, which make it a superior material for the group velocity delay compensation in the femtosecond pulse application. BBSAG is a new nonlinear crystal invented by Chen et al. [16]. It is an optimized BBO crystal with larger SH generation efficiency and higher optical threshold. Rui Wang et al. [18] reported a single-longitudinal-mode DUV laser through frequency quadrupling of a tunable Ti:Sapphire amplifier system, in which BBSAG was employed in the final SFM stage. Tunable DUV pulse was obtained in the range of 195–205 nm, with a maximum output power up to 130 mW.

In this letter, we have demonstrated a high efficient frequency tripling with collinear group velocity compensation for wavelength-tunable ultrafast UV laser generation. A new nonlinear crystal BBSAG is used in the SH generation stage for high efficiency conversion. A 3-mm-long BBO crystal cut at 65° is used as a tunable group velocity compensation plate in a collinear SFM scheme. A factor of 4.4 times improvement in the TH generation efficiency has been obtained compared with the collinear SFM without compensation. Tunable femtosecond UV pulses in the wavelength range from 256.7 to 276.7 nm are generated by accurate adjustment of crystal angles, with a maximum power of 212 mW, corresponding to single pulse energy up to 0.21 mJ.

2. Materials and methods

BBSAG is a kind of low-temperature phase barium metaborate single crystal belonged to trigonal system, doped with one or more elements selected from Si, Al and Ga. As an optimized BBO crystal, BBSAG completely overcomes the shortcoming of deliquescence compared with BBO [18], and its nonlinear efficiency and optical damage threshold has also been improved greatly. For these reasons we use BBSAG to generate the SH pulse in the first stage.

In the femtosecond regime, short pulse provides high efficient frequency conversion due to its high peak powers, but the GVM is a cognitive factor to limit the conversion efficiency. Owing to the refractive index difference between the fundamental and SH pulse in the crystal, the group velocity is quite different, as shown in Fig. 1. This difference causes the two pulses to separate in time with a time delay Δt, which is given by:

$$\Delta t = L_{SHG}\left(\frac{1}{V_F} - \frac{1}{V_{SHG}}\right) \quad (1)$$

where V_F and V_{SHG} denote the corresponding group velocities of the fundamental and the SH pulse, respectively, and L_{SHG} is the crystal length. According to expression (1), the time delay (Δt) from a 1-mm-long BBSAG crystal (for type-I phase matching) is about −193.3 fs at 800 nm, as shown in Fig. 2. It is obvious that the SH and fundamental pulses separate from each other after the frequency doubling. The SH pulse lags behind the fundamental as

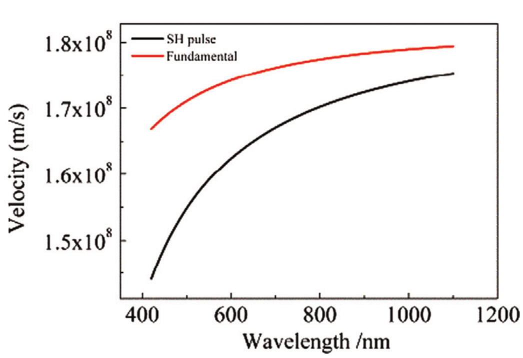

Fig. 1. Group velocity of fundamental and SH pulses in BBSAG crystal.

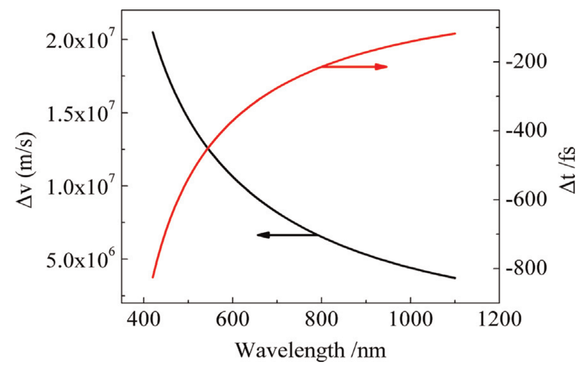

Fig. 2. Group velocity mismatch and time delay between fundamental and SH pulse (SH pulse lags behind the fundamental).

they propagate through the crystal and it is broadened to longer pulse duration than the fundamental pulse. It is reasonable to use type-II phase matching for TH generation, since the SH pulse is polarized perpendicular to the fundamental. The TH pulse is generated only by the overlap of the part of the fundamental and SH owing to the high group velocity difference, which makes the conversion efficiency of the TH generation less than perfect. We expect the group velocity walk-off as little as possible, so a very thin crystal suits for requirement. Unfortunately, the conversion efficiency of SH generation will decrease with very thin crystal. Usually the nonlinear crystal length is chosen to be shorter than the shortest temporal walk-off length of the interacting pulse. Considering the effect on both group velocity walk-off and conversion efficiency of SH, a 1-mm-long BBSAG crystal is used for SH generation. Another advantage is the broadening of SH pulse is little.

The conventional method to compensate the GVM is to adjust the path length between the fundamental and SH pulse by means of time delay line which consists of beam splitters and mirrors. This design requires that the two optical path lengths must be exactly the same and the length should be adjusted carefully. Another drawback of time delay line is that the energy of fundamental and SH pulses introduces severe losses by getting through the beam splitters and mirrors. It also makes the experimental setup complex. In this paper, as shown in Fig. 3, we use a tunable collinear group velocity compensation plate to correct the time delay between the SH and TH generation crystals. The compensation plate made of birefringent crystal is cut with the optical axis oriented in a suitable angle. The fundamental pulse lags behind SH in the compensation plate. BBO crystal suits for requirement owning to large birefringence and acceptance bandwidth. An additional advantage of BBO is the high threshold and good surface quality. We calculate two different cut angle of BBO crystals, as shown in Fig. 4. The time delay is 318 fs and 211 fs in 3-mm-long, θ=70°-cut BBO (φ=0°) and 3-mm-long, θ=65°-cut BBO (φ=0°), respectively, when the angle of incidence is close to 0°. According to prior calculation, the delay from SH generation crystal is −193 fs. Moreover, it is a good idea to pre-compensate a certain amount of extra time delay between the fundamental and SH pulse because the TH generation crystal also has time delay. The TH pulses are produced in a 0.5-mm-long type-II BBO crystal. This

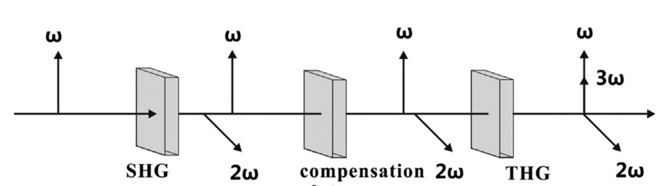

Fig. 3. Generation of TH with collinear group velocity compensation technique.