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## Generation of high beam quality, high-energy and broadband tunable mid-infrared pulse from a KTA optical parametric amplifier



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#### 1. Introduction

Few-cycle 3-5 µm femtosecond laser sources in the mid-infrared field are essential for many applications including timeresolved spectroscopy, strong-field laser-matter interactions and so on. So far, high intensity and high-power mid-infrared pulses have been used for exploring the strong-field ionization, because electrons acceleration occurs in a longer time interval and obtains higher collision energy with a long wavelength at a given intensity [1–3]. Another direct application of mid-infrared laser pulse in strong-field physics can be found to drive high harmonic generation (HHG), because mid-infrared pulses possess the ability to increase the cut-off energy [4-7]. In addition, the tunable highenergy mid-infrared sources have also been used for glycol-protein structure analysis and demonstrated photo-dissociation of specific bonds in a glycol-protein molecule [8]. The vibrationalrotational energy of large glycol-peptide molecules is corresponding to the photon energy of the mid-infrared coherent light sources. Furthermore, with the advantage of high temporal resolution and sensitivity, the mid-infrared lasers are excellent sources for remote sensing and nanoimaging [9,10].

Based on the demands of the mentioned and potential applications, it is necessary to develop high-energy mid-infrared laser sources with broadband tunability. Because the lack of suitable conventional gain media in the mid-infrared field, nonlinear frequency conversion has been a typical way to generate ultrashort

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#### ABSTRACT

We have demonstrated efficient generation of high beam quality, high-energy and broadband tunable femtosecond mid-infrared pulses using a three-stage collinear optical parametric amplifier (OPA). The white-light continuum (WLC) seeded OPA setup, based on KTA crystal in three stages and pumped by a femtosecond laser pulse at 800 nm, is capable of producing idler wavelength ranging from 2.4  $\mu$ m to 4.0  $\mu$ m with energy up to 82  $\mu$ J at 3.27  $\mu$ m, which corresponds to signal energy of 350  $\mu$ J at 1060 nm. The output pulse has excellent intensity distribution with measured beam quality factor  $M^2 \sim 1.1$  for signal and  $M^2 \sim 1.7$  for idler. To our knowledge, this is the best beam quality reported in 3–5  $\mu$ m femtosecond OPA until now. The achieved mid-infrared pulse also has a good energy stability with a fluctuation of 1.01% rms over half an hour.

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mid-infrared laser pulses. The typical and efficient approach of nonlinear frequency conversion to generate tunable mid-infrared pulses is nonlinear frequency conversion, such as optical parametric amplification (OPA), optical parametric oscillator (OPO) and optical parametric chirped pulse amplification (OPCPA).OPO can be used for generating broadband and high repetition rate midinfrared pulses, but it is limited by the low output energy and the bandwidth of the mirror coatings and it also must overcome the problem of timing-jitter which comes from the quantum-noise seed [11-14]. OPCPA can also generate high peak power pulses, however it is complex and expensive for the stretcher and compressor, and the delay control and idler compression can be challenging [15–18]. OPA seeded with a white-light continuum has the advantage of high single-pass gain, broad tuning band, straightforward passive carrier-envelope-phase (CEP) control and is simpler to operate, thus OPA is very compact and efficient, and also it can be straightforwardly used as a building block for the socalled "perfect waveform" with no post compression, so it is better to use OPA to generate mid-infrared laser pulses. Many reports have been demonstrated experimentally [19-22]. Such as 2-µJ pulses tunable in the 3-4  $\mu$ m range with bandwidth supporting 30-fs transform-limited duration by using LiLO<sub>3</sub> crystal has been demonstrated by D.Brida et al. in 2007 [23], but the pulses energy was guite low. Tobias Steinle and Andy Steinmann have reported a Watt-level OPA at 42 MHz with wavelength tuning from 1.35 to 4.5 µm coherently seed with solitons [24]. The repetition was very high, but the pulses energy was still quite low and the beam quality was not reported. Magnus W. Haakestad and Gunnar Arisholm have obtained high energy mid-infrared laser source in the 3–5  $\mu$ m with  $M^2$ =2–4 in a ZGP OPA pumped by a 6 ns Q-switched Nd:YAG laser, the system is relatively complex with two OPOs and two OPAs [25]. As we know, beam quality is one of the most important parameters among the beam characteristics, and it has a great influence on the energy distribution, focusing property and power density. A good beam quality is the fundament of the beam propagating over a long distance with no distortion. Until now, it is still a great challenge to obtain high pulse energy few-cycle  $3-5 \,\mu\text{m}$  femtosecond laser sources with good beam quality.

In recent years, with the rapid development of laser technology, such as kHz Ti:sapphire laser, it greatly facilitates the generation of WLC seed with an excellent spatial quality and a very high pulse stability in a variety of materials [26], such as YAG, Al<sub>2</sub>O<sub>3</sub>, and it has become one of the most popular ultrafast laser sources for mid-infrared OPA systems [27–29]. Moreover, with the development and gradually maturity of the nonlinear crystals technology in the mid-infrared field, nonlinear process is playing an important role in the field of ultrashort mid-infrared laser sources.

In this paper we report on a collinear OPA system for high beam quality and broadband tunable mid-infrared femtosecond laser pulse generation pumped by a Ti:sapphire laser amplifier at 800 nm. The tunability range of the idler pulse covering 2.4–4.0 µm is demonstrated with a three-stage KTA femtosecond OPA, with energy up to 82µJ at 3.27 µm. The maximum energy net conversion efficiency can reach 14.6%. The beam quality is perfect corresponding to the quality factor  $M^2 \sim 1.1$  for signal and  $M^2 \sim 1.7$  for idler, respectively. And the achieved mid-infrared pulse has a good energy stability with a fluctuation of 1.01% rms over half an hour.

#### 2. Theoretical analysis

As we know, nonlinear crystal is a key in the OPA process. High transmission, relative high damage threshold, high nonlinear coefficient in the mid-infrared wavelength range is important. Such as KTA crystal is a kind of positive biaxial crystal belonging to 2-mm point group, is transparent in a broad wavelength of 0.4–5  $\mu$ m and it has a high nonlinear coefficient of > 3 pm/V, no absorption at 3.5  $\mu$ m. Also, it has a very high damage threshold of > 200 GW/ cm<sup>2</sup> pumped by 800 nm with a pulse duration of 200 fs, and a high thermal conductivity of about 20 W/(*m*\**K*), which can avoid the obvious thermal lens effect under a strong pump intensity [30,31]. So, KTA crystal is an ideal material in the 3–5  $\mu$ m research field.

According to its refractive index Sellmeier's equations, type II phase matching condition  $(o_i + e_s - > o_p)$  can be satisfied, and the most efficient conversion is obtained in the x-z plane. We calculate the phase matching condition  $(o_i + e_s - > o_p)$  pumped with Ti:sapphire laser centered at 800 nm, and the calculated phase matching tuning cure is shown in Fig. 1. The phase matching angle doesnot change too much as the signal wavelength varies from 1000 to 1200 nm, which indicates that it is capable of providing broadband tunable pulses. Moreover, femtosecond pulses propagate in the KTA crystal with different group velocities  $v_g = \frac{d\omega}{dk}$ , the group velocity mismatch (GVM) between the pump and the signal/ idler pulse ( $\delta_{pj} = \frac{1}{v_{gp}} - \frac{1}{v_{gj}}, j = s, i$ ) limits the effective interaction length. Fig. 2 shows the GVM curves for type II phase matching of KTA crystal under 800 nm pump. For the signal in the wavelength range of 1000–1060 nm, we can find that it satisfy  $\delta_{vs}\delta_{vi} < 0$ , thus the signal and idler pulse propagate in opposite direction with respect to the pump pulse. In this way, they tend to stay localized under the pump pulse, and this effect increases the interaction length and presents an exponential gain for amplification.



Fig. 1. Phase matching tuning curve for KTA OPA at the pump wavelength of 800 nm for type II.



**Fig. 2.** Pump –signal ( $\delta_{ps}$ ), pump-idler ( $\delta_{pi}$ ) and signal-idler ( $\delta_{si}$ ) group velocity mismatch curves for KTA OPA at the pump wavelength of 800 nm for type II.

Especially, the largest gain and highest output energy should be obtained when the group velocities of the pump and the signal are matched at the signal wavelength of 1060 nm (corresponding to the idler wavelength of 3.26 µm) theoretically. In addition, the GVM between signal and idler pulse limits the phase matching bandwidth. According to  $\Delta \nu \propto \frac{1}{\left|\frac{1}{\nu g_{S}} - \frac{1}{\nu g_{S}}\right|} = \frac{1}{|\delta_{SI}|} [26]$ , the gain band-

width dramatically decreases when the GVM between signal and idler is increasing. In this case, the gain bandwidth of the signal near 1000 nm is about twice larger than that near 1200 nm. Thus the idler in the range of  $3.5-4 \,\mu\text{m}$  is in a broadband interaction region for amplification with a larger gain bandwidth compared to that shorter than  $3.0 \,\mu\text{m}$  in a narrowband region.

#### 3. Experimental setup

The experimental setup of the mid-infrared OPA system is schematically shown in Fig. 3. This system is pumped with a commercially available femtosecond Ti:sapphire laser centered at 800 nm with 1 kHz repetition (Coherent, Astrella). Energy of 2.95mJ is sent into the OPA system as the pump. Our system configuration consists of a pre-amplifier (OPA1) and two power amplifiers (OPA2 and OPA3). The three nonlinear KTA crystals are

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