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Ultra-broadband optical parametric amplification in $KBe_2BO_3F_2$ crystal

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Abstract

It has been demonstrated for the first time that ultra-broadband phase-matching coupled with group-velocity matching is possible in a type-I non-collinear optical parametric amplifier in a recently grown $KBe_2BO_3F_2$ (KBBF) crystal, pumped by Ti:sapphire second harmonic 395 nm radiation and seeded by white-light continuum.

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Keywords: Optical parametric amplification; New NLO crystal; Ultrashort pulse generation; KBBF

1. Introduction

Due to the unique structural characteristics, borate group nonlinear optical (NLO) crystals have been found to be very useful in various NLO applications, including in femtosecond experiments. Among other crystals of this family, BBO has been proved as a potential material in ultrafast NLO research applications [1–5]. Utilizing the ultra-broadband phase-matching (UBPM) property of BBO crystal, first discovered by Gale *et al.* [1], several interesting research reports have come out and the pulse-width of the tunable visible-near infrared (vis-NIR) optical pulse has now been reduced to as short as ~4 fs in the visible using type-I non-collinear optical parametric amplification (NOPA) [4]. Recently several potential borate group crystals, including $CsLiB_6O_{10}$ (CLBO), $K_2Al_2B_2O_7$ (KABO), $Li_2B_4O_7$ (LB4), and $KBe_2BO_3F_2$ (KBBF) have been discovered with improved NLO properties [6–9]. NLO crystals being in the heart of NOPA, it is always important to search the potentialities of these newly developed crystals for use in different NOPA experiments.

In earlier reports [6,7], we have demonstrated the potentialities of several newly developed potential borate group crystals, such as BBO, CLBO, KABO, and LB4 crystals for generation of ultrashort optical pulses by NOPA. Another borate based NLO crystal, KBBF has recently been developed and it has been proved [8,9] that KBBF is the best NLO crystal to achieve deep and even vacuum UV second harmonic generation up to now. However, here we present for the first time the ultra-broadband phase-matching property of the KBBF crystal for generation of ultrafast laser radiation by type-I NOPA pumped by Ti:sapphire second harmonic (395 nm) radiation and seeded by white-light continuum.

2. Experimental arrangement

Fig. 1a shows the schematic of the non-collinear interaction geometry to be employed in experiment for achieving

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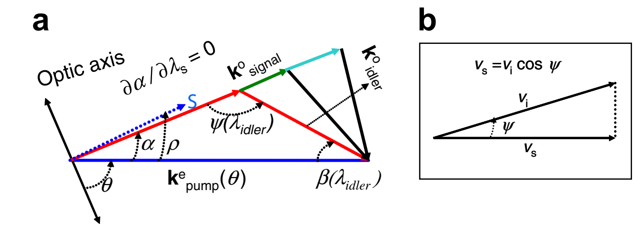

Fig. 1. (a) Schematic of the nonlinear optical interaction geometry for achieving broadband amplification through non-collinear optical parametric amplification (NOPA). (b) Group-velocity matching condition in type-I NOPA.

the generation of efficient and broadband ultrashort laser radiation through type-I NOPA in KBBF crystal. Here, the pump radiation is the Ti:sapphire second harmonic radiation, i.e. 395 nm and seed radiation is the white light continuum (WLC) generated in a sapphire plate pumped by the same 395 nm radiation. The Ti:sapphire fundamental radiation may also be used as the pump for WLC generation. As per the requirements of type-I interaction in KBBF crystal, the pump radiation is e-polarized and the signal and the idler radiations are o-polarized. The symbols $k_p^e(\theta), k_s^o$, and k_i^o of Fig. 1a are the wave vectors for the e-polarized pump, o-polarized signal, and o-polarized idler radiations, respectively. The non-collinear angle between the pump and signal (idler) is designated as $\alpha(\beta)$, and that between the signal and idler beams is designated as ψ, and ρ is the walk-off angle. The phase-matching angle θ is defined as the angle between the pump radiation and the optic axis direction of the crystal, unless specified otherwise. As shown in Fig. 1, that the propagation direction of the seed radiation inside the crystal must be in the direction of the spatial walk-off ρ of the e-polarized pump radiation to alleviate the deleterious walk-off effect. The calculated value of ρ and the optimum non-collinear angle (α_{opt}) for broadband phase-matching is the same and it is 2.5°. This property of KBBF crystal is very much advantageous for achieving good conversion efficiency in the employed NLO interaction. The method of calculation of θ and α_{opt} have been elaborated later.

3. Group-velocity matching and phase-matching in KBBF

It has been demonstrated previously by several authors [1–7] that for achieving broadband phase-matching in a type-I NOPA it is required to satisfy the conditions of group-velocity matching and phase-matching simultaneously. Among others, the inverse group-velocity mismatch ($GVM_{s–i}$) between the signal and idler radiations of a NOPA is an important parameter as this determines predominantly the bandwidth of a NOPA [6]. Here, in Fig. 1b, the condition of group-velocity matching is illustrated. The value of $GVM_{s–i}$ can be calculated using the following equation.

$$GVM_{s\text{–}i} = 1/v_s - 1/(v_i \cos\psi), \quad (1)$$

where v_s and v_i are the group velocities for the signal and idler radiations, respectively. From a practical viewpoint it is useful to calculate the optimal value of the pump-signal non-collinear angle (α_{opt}) that corresponds to the zero value of $GVM_{s–i}$. The value of $\alpha_{opt,}$ in terms of other parameters, such as the refractive indices (n_s, n_i), interacting wavelengths (λ_t, λ_s) and non-collinear angle (ψ) between the signal and idler beams, can be calculated as [6]

$$\alpha_{opt} = \sin^{-1}[(1-(v_s/v_i)^2)/(1+(n_s/n_i)^2(\lambda_i/\lambda_s)^2 + 2(n_s/n_i)(\lambda_i/\lambda_s)(v_s/v_i))]^{1/2}. \quad (2)$$

The variation of α_{opt} with the signal wavelength is demonstrated in Fig. 2. By employing suitable non-collinear geometry ($\alpha = \alpha_{opt}$) group-velocity matching is possible for a wide signal wavelength region unlike for collinear case where group-velocity matching is allowed only in the degeneracy condition as is observed in Fig. 3, for 395 nm pumped type-I KBBF NOPA.

Fig. 4 shows the phase-matching characteristics of 395 nm pumped type-I NOPA with a KBBF crystal as a

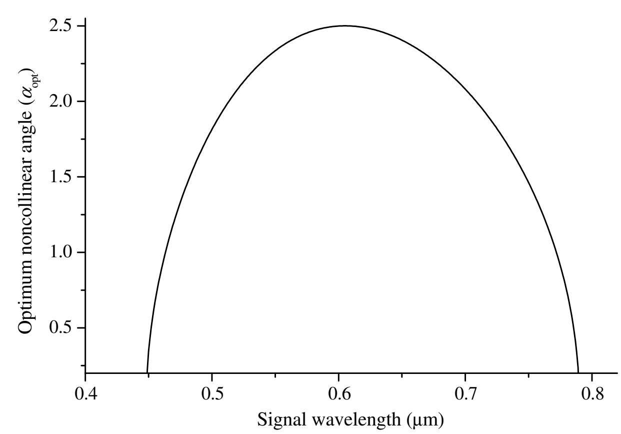

Fig. 2. Variation of the optimum non-collinear angle (α_{opt}) with the signal wavelength of 395 nm pumped type-I NOPA in KBBF crystal.

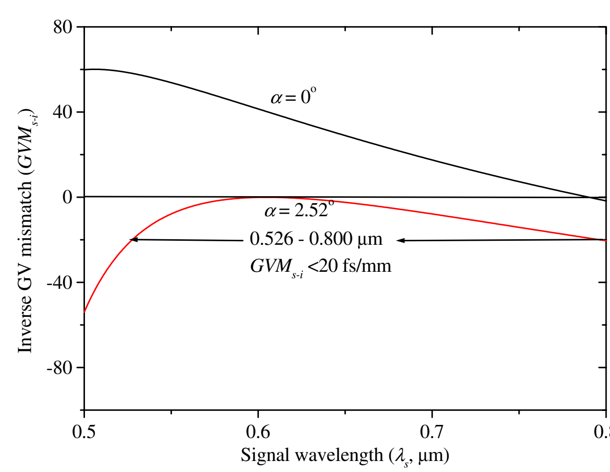

Fig. 3. Group-velocity matching in type-I KBBF NOPA pumped by Ti:sapphire second harmonic 395 nm radiation.

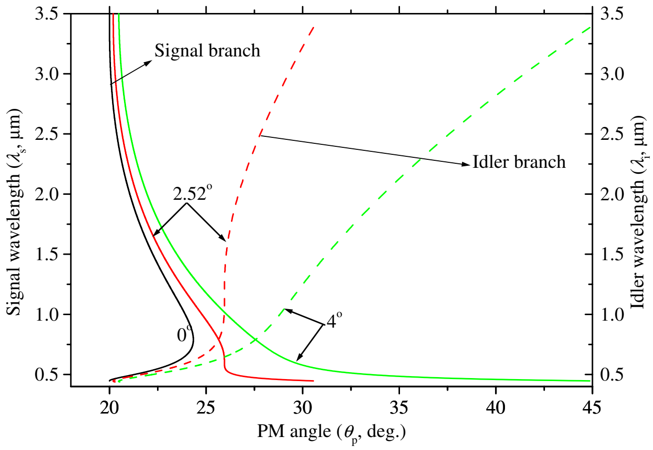

Fig. 4. Phase-matching characteristics (signal and idler branches) of type-I KBBF NOPA for $\alpha = 0°$, 2.52°, and 4.0°. Here pump = 395 nm.

gain medium. Calculation was made for three different non-collinear angles values of $\alpha = 0°$, 2.52°, and 4.0° covering the entire tuning range (0.45–3.5 μm). Fig. 5 shows the phase-matching characteristics covering only the region 0.45–0.80 μm and in this graph we have plotted the variation of pump (θ_p (or θ)) signal (θ_s) and idler (θ_i) propagation directions inside the crystal when $\alpha = 2.52°$. It is found from Fig. 5 that the ultrabroadband phase-matching is possible in type-I KBBF NOPA for this value of α. Here, θ_p (or θ), θ_s, and θ_i are defined as the angle made by the pump (λ_p), signal (λ_s), and idler (λ_i) radiations, respectively, with the optic axis of the crystal. The phase-matching angles have been calculated by using the following equation, which is obtained from the energy and momentum conservation principles [6]

$$\theta = \cos^{-1}\left[\frac{(k_p^e/k)^2 - 1}{(k_p^e/k_p^o)^2 - 1}\right]^{0.5}, \quad (3)$$

where, $k = \sqrt{[(k_s^o)^2 + (k_i^o)^2 + 2k_s^o k_i^o \cos\psi]}$, k may be defined as "averaged" wave vector of signal and idler along the pump direction. From the calculation results it is found that for a particular value of $\alpha = 2.52°$ the phase-matching curves for pump (and also for signal) is flat, viz., for the variation of λ_s in the region 0.534–0.758 μm θ_p as well as θ_s varies within $25.9 \pm 0.1°$, and so generation of broadband amplified signal radiation lying between 0.45 and 0.80 μm is possible by NOPA with a KBBF crystal.

Fig. 6 shows the calculated approximate efficiency of the parametric amplification process in a 1-mm long KBBF crystal pumped by 395 nm, for $\alpha = 2.52°$. The parametric efficiency has been calculated using infinite plane-wave approximation and it is proportional to $\mathrm{sinc}^2(\Delta kL/2)$ function, where $\Delta k = k_p^e(\theta_p) - k_s^o\cos\alpha - k_i^o\cos\beta$, and L is the crystal length. From the Fig. 6 it is observed that the full width of the curve at half maximum (FWHM) level of parametric efficiency is 217 nm centered at 620 nm. During the calculations the refractive indices of KBBF crystal were calculated using the following Sellmeier dispersion relations of Chen *et al.* [8]:

$$n_o^2 = 1 + 1.169725\lambda^2/(\lambda^2 - 0.0062400) - 0.009904\lambda^2, \quad (4a)$$

$$n_e^2 = 1 + 0.956611\lambda^2/(\lambda^2 - 0.0061926) - 0.027849\lambda^2, \quad (4b)$$

with λ in μm.

Broadband amplification of radiation is possible through type-I NOPA in several other newly developed crystals, such as BBO, CLBO, KABO, and LB4 [6,7]. Therefore, here we have compared different linear and nonlinear optical properties of KBBF crystal with those of BBO, CLBO, KABO, and LB4 crystals and the results are summarized in Table 1. The optical transmission range of KBBF crystal as shown in Table 1 is the shortest in the VUV down to 155 nm, which is very much advantageous in the device point of view. Another parameter of importance of a NLO crystal used in a type-I NOPA is its walk-off angle (ρ) at the pump wavelength. The walk-off effect is detrimental to achieve large gain in an optical parametric process as it reduces the effective interaction length of the

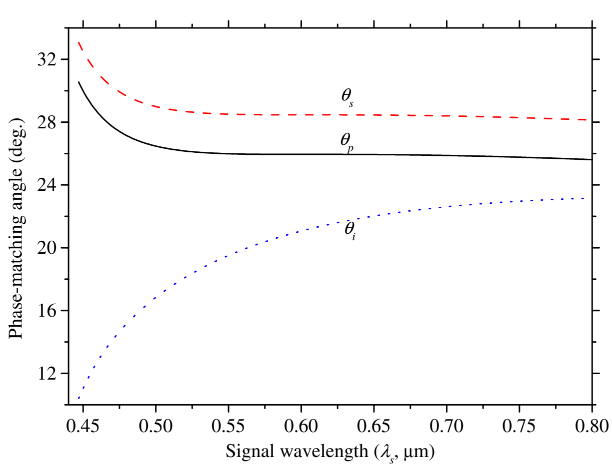

Fig. 5. Ultra-broadband phase-matching in type-I KBBF NOPA with $\lambda_p = 395$ nm and $\alpha = 2.52°$. Here, θ_p, θ_s, and θ_i are pump, signal and idler phase-matching angles, respectively.

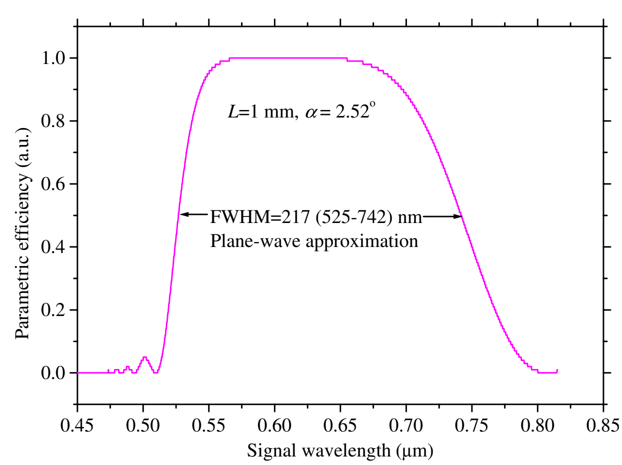

Fig. 6. The efficiency of broad-bandwidth parametric amplification, in KBBF type-I NOPA for 395 nm pump wavelength, in the plane-wave approximation.

Table 1
Summary of NLO parameters of BBO, CLBO, KABO, LB4, and KBBF crystals for type-I NOPA

NLO crystal	Transmission range (nm)	ρ (°)[a]	α_{opt} (deg)[b]	Bandwidth (THz)[c]	NLO coeff. (pm/V)	LDT (GW/cm^2)[d]	Chemical stability	Commercial availability
BBO	188–3300	4.0	3.7	157	$d_{22} = 2.3$	~13	Hygroscopic	Available
KABO	180–3600	2.7	3.4	152	$d_{11} = 0.45$	>15	Non-hygroscopic	Not available
CLBO	175–275	2.1	3.0	164	$d_{36} = 0.95$	~26	High-hygroscopic	Available
LB4	160–3500	2.1	2.9	174	$d_{31} = 0.15$	~40	Non-hygroscopic	Available
KBBF	155–3660	2.5	2.5	174	$d_{11} = 0.76$	≫40	Non-hygroscopic	Not available

[a] Spatial walk-off angle at the pump wavelength of 395 nm.
[b] Optimum non-collinear angle for $GVM_{S-I} = 0$, for the signal wavelength ~600 nm.
[c] FWHM of the acceptable power spectrum of NOPA fot $L = 1$ nm.
[d] At Nd:YAG 1064 nm wavelength with ns pulse.

crystal. However, by judicious choice of non-collinear configurations, as shown in Fig. 1a, it is possible to overcome the deleterious effect of the walk-off. In the case of BBO crystal ρ is almost equals to the optimum non-collinear angle (α_{opt}) required to achieve broadband phase-matching and this fortuitous matching is made possible in BBO to achieve large gain as well as large bandwidth [1–5]. From the results as summarized in Table 1, it is found that for KBBF crystal the value of α_{opt} as well as that of ρ is ~2.5°. Therefore, by employing non-collinear geometry it is possible to alleviate the deleterious walk-off effect in type-I KBBF NOPA and achieve the large gain as well as wideband amplification also in KBBF crystal that was exploited earlier in BBO crystals [1–5]. The nonlinear optical coupling coefficient (d_{eff}) of KBBF type-I NOPA has been compared with those of BBO, KABO, CLBO, and LB4 crystals. From the Table 1 it is observed that the value of d_{eff} of KBBF crystal is larger than those for LB4 and KABO crystals, however, smaller than those of BBO and CLBO crystals. However, the disadvantage of low value of d_{eff} can be overcome by employing tight focusing of the incident radiations as the laser damage threshold (LDT) of KBBF is largest in comparison with those of the other borate crystals. However, considering that the parametric gain (G) of the NOPA is approximately proportional to $d_{eff}^2 I_p L^2$, the damage threshold intensity of KBBF is >3 times that of BBO and d_{eff} is ~1/3th to that of BBO, with the highest pump intensity (I_p) the maximum value of G of KBBF NOPA that can be achieved should be larger or at least comparable to that of BBO if the crystal lengths are same.

4. Conclusions

In conclusion here we have presented for the first time the NLO properties of the recently discovered KBBF crystals for generation of short pulse vis-NIR laser radiation by type-I NOPA pumped by 395 nm radiation. It is found that for a particular value of $\alpha = 2.52°$ phase-matching curve is flat and using a 25.9° type-I cut KBBF crystal generation of ultrafast laser radiation having bandwidth >217 nm, centered at 620 nm is possible. Generation of laser pulses with pulse duration as short 4-fs have been reported in BBO through NOPA [4]. Also by employing the technique presented in this work, generation of laser radiation with sub 4-fs pulse duration containing less than two optical cycles is possible in KBBF crystals. However, proper care is to be taken for the chirp compression [4,5] of the generated broadband radiation as the seed pulse (white light continuum) to be used in such experiments is chirped. KBBF crystal has some other advantageous characteristics, such as a high damage threshold, non-hygroscopic in nature, and shortest SHG cut-off [8,9]. However, due to difficulties in growing, the KBBF crystal sample of proper cutting angle is yet to be available for commercial uses.

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