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Nonlinear optical properties of $Li_2B_4O_7$ (LB4) crystal for the generation of tunable ultra-fast laser radiation by optical parametric amplification

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ABSTRACT Using a very simple and straightforward approach we derive the condition to be satisfied for achieving wavelength-insensitive broadband phase matching in a type-I noncollinear optical parametric amplifier (NOPA), required for the generation of ultra-fast laser radiation. Nonlinear optical properties of a relatively newly grown crystal $Li_2B_4O_7$ (LB4) have also been studied and we found that this crystal satisfies the condition required to realize the broadband phase matching and is suited for the generation of tunable visible–near-infrared ultra-fast laser radiation employed in a 395-nm-pumped type-I NOPA. The phase-matching bandwidths of type-I NOPAs in different borate-group crystals, such as BBO, CLBO, and KABO, are also numerically estimated. The values are ~ 157, 164, 152, and 174 THz for 1-mm-thick BBO, CLBO, KABO, and LB4 crystals, with the noncollinear angles between the input pump and the signal beams 3.7°, 3.0°, 3.4°, and 2.9°, respectively, for the signal wavelengths centered at 630 nm. In addition to the largest bandwidth, LB4 crystal has several other attractive properties to be used in optical parametric applications, such as high laser damage threshold, wide optical transmission, easy crystal growth to excellent optical quality with large sizes, easy treatment of cutting and polishing, and nonhygroscopicity.

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1 Introduction

The recent progress in the development of solid-state laser materials along with nonlinear optical (NLO) materials has brought a resurgence of interest in the development of tunable ultra-fast laser radiation sources employing the optical parametric amplification technique. Ultra-fast pulses have ultra-broad bandwidth and so for the generation of ultra-fast pulses it is required to generate ultra-broadband radiation. This becomes possible by employing the optical parametric amplification technique where a broadband seed pulse is amplified at the expense of the energy of the intense pump radiation [1–10]. For a fixed value of crystal orientation and temperature, the bandwidth of an optical parametric device is restricted by the law of momentum conservation or equivalently by phase matching, and in general it is satisfied for a single set of wavelengths of the pump, signal, and idler radiations. However, in some type-I phase-matched parametric devices it is possible to achieve a broad bandwidth by realizing a first-derivative insensitivity of the phase-matching angle to the signal wavelengths by employing noncollinear geometry [1–10]. The condition to achieve the insensitivity of the phase-matching angle to the signal wavelengths in an optical parametric oscillator was mentioned first by Hache et al. [2] and then employed by several researchers using BBO as NLO material in their noncollinear optical parametric amplifiers (NOPAs) [1–10]. However, we derive the same condition to achieve the insensitivity of the phase-matching angle to the signal wavelengths of a type-I NOPA, using a straightforward approach, starting from an expression of the phase-matching angle. The derivation presented here carries its own significance due to its simplicity and also the physical meaning of the equations involved can be easily understood.

In recent years several potential borate-group crystals, such as $CsLiB_6O_{10}$ (CLBO) and $K_2Al_2B_2O_7$ (KABO), have been discovered to have potentialities for different NLO applications particularly for the generation of UV/VUV laser radiation [11–19]. Recently, we studied optical parametric properties of these crystals and found that they have attractive properties similar to those of BBO crystal and are also suitable for the generation of tunable ultra-fast laser radiation by employing the type-I noncollinear optical parametric amplification technique [20]. Another borate-group NLO crystal, namely $Li_2B_4O_7$ (LB4), has been successfully grown in the recent past by Komatsu et al. by the Czochralski method [21]. This crystal possesses several favorable properties to be used in practical parametric and other NLO frequency-mixing devices. It is a negative uniaxial crystal and has clear transmission spans in the wavelength range extending from 170 to 3300 nm; however thinner samples transmit even down to 160 nm (i.e. VUV cut-off of this crystal is ~ 29 nm below that of BBO and ~ 20 nm below that of CLBO and KABO crystals) as well as in the near infrared (NIR) up to ~ 3500 nm. Having a wider band gap the LB4 crystal has a high laser damage threshold of about 40 GW/cm^2 at 1064 nm and 10-Hz repetition rate with 10-ns laser radiation [21]. The value of the damage threshold of LB4 crystal is about three times

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**On leave from:* The Department of Physics, Regional Engineering College, Durgapur-713209, Burdwan, India that of BBO crystal and about 1.5 times that of CLBO crystal. Some other attractive advantages are the following. It is mechanically rigid and nonhygroscopic, whereas BBO and CLBO crystals are soft and hygroscopic. CLBO crystal is especially highly hygroscopic and requires maintenance of specific atmospheric conditions such as temperature and humidity. The deleterious extraordinary beam walk-off in LB4 crystal is smaller than in BBO and KABO crystals and similar to that in CLBO crystal, viz. at the pump wavelength (395 nm) of a NOPA, the walk-off angles are about 4.0°, 2.7°, 2.1°, and 2.1° in BBO, KABO, CLBO, and LB4 crystals, respectively. High-quality LB4 crystals can be grown with much larger sizes than those of other crystals of the borate family, such as BBO, CLBO, and KABO. Although the LB4, BBO, and CLBO crystals are available commercially, the growth process of KABO crystal has been developed very recently [17, 18] and it is yet to be available commercially. The only disadvantage of the LB4 crystal is that its effective nonlinear coefficient is somewhat small ($d_{31} = 0.15$ pm/V at 1064 nm) [22]. The value of d_{31} was later corrected to be 0.21 pm/V [23]. However, the disadvantage of low nonlinearity may be overcome at least partially by its high value of the laser damage threshold employing tightly focused geometry in NLO interactions. The potentiality of this crystal had been proved for the generation of tunable ns pulsed UV, VUV, and NIR laser radiation through second-harmonic generation, sum-frequency mixing, and difference-frequency mixing processes, respectively, using Q-switched Nd : YAG-laser-based fundamental laser radiation sources [21, 22, 24–26]. In [23], the potentiality of this crystal had been proved for its femtosecond VUV applications, where the authors successfully generated VUV pulses down to 170 nm using the sum-frequency mixing technique. However, no effort has been made so far to study the potentiality of this newly developed borate crystal for its optical parametric applications.

Here we study the nonlinear optical properties of this crystal when it is used in a type-I NOPA pumped by Ti : sapphire second-harmonic radiation at 395 nm and seeded by the white-light continuum (WLC). From the study it is found that this crystal also satisfies the condition required for broadband phase matching when a suitable noncollinear angle between the input pump and seed pulses is considered. The numerically estimated value of the bandwidth of the type-I LB4 NOPA is > 174 THz when an internal noncollinear angle (α) of 2.9° is employed for a 1-mm-thick crystal. For some other borate crystals, such as BBO, KABO, and CLBO, the values of the phase-matching bandwidths for the same interaction and also for the same thickness have also been calculated and the values are over 157, 152, and 164 THz, with the values of α being 3.7°, 3.4°, and 3.0° for BBO, KABO, and CLBO crystals, respectively. Considering the different advantageous characteristics, such as easy growth to good-quality, large-size crystals, short VUV cut-off wavelength, high value of laser damage threshold, small walk-off angle, easy treatment of cutting and polishing, and nonhygroscopic nature, LB4 crystal might be an alternative crystal for the generation of tunable visible–NIR ultra-short laser radiation by employing a 395-nm-pumped type-I NOPA. However, we present in this article the optical parametric properties of LB4 crystal for the first time, to the best of our knowledge, and found that, in principle, the generation of tunable ultra-fast laser radiation is possible using this crystal in a type-I NOPA pumped by Ti : sapphire second-harmonic radiation and seeded by the WLC, with the bandwidth largest among other borate crystals such as BBO, KABO, and CLBO.

2 Condition for signal-wavelength-insensitive phase matching in a negative uniaxial crystal employed in a type-I NOPA

For the generation of ultra-fast laser radiation the generation of ultra-broadband radiation is required and the optical parametric processes have now become the well-established techniques for the generation of such widely tunable laser radiation. For the generation of ultra-broadband laser radiation in a type-I NOPA the orientation and temperature of the NLO crystal are kept fixed, and so in general the bandwidth of the parametric process is limited by the momentum conservation or phase matching to a single set of frequencies of the pump, signal, and idler radiations. However, if the NLO crystal satisfies a certain condition for the first-derivative insensitivity of the phase-matching angle with respect to the signal wavelength of the type-I NOPA, it is possible to achieve the broad bandwidth. A derivation of the above condition in a straightforward manner is presented below.

The standard method of calculation for the phase-matching angle for different NLO applications in different NLO crystals is available in the literature [27]. In addition, recently a very simple expression of the phase-matching angle for different collinear and noncollinear type-I NLO interactions in a negative uniaxial crystal was presented [20]. For the sake of completeness some of the results are also presented below. Figure 1a shows the noncollinear geometry and Fig. 1b shows the group-velocity-matching condition between the signal

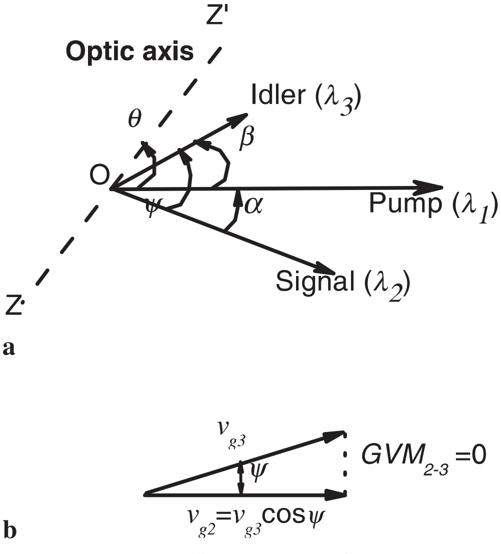

FIGURE 1 **a** Noncollinear geometry for type-I NOPA. The internal noncollinear angles between the pump and the signal (idler) and between the signal and the idler radiations are α (β) and ψ, respectively. **b** The condition for group-velocity matching between the signal and the idler (GVM$_{2-3}$) radiations of the NOPA. Here ν_{g2} and ν_{g3} are the group velocities of the signal and the idler radiations, respectively

and idler radiations of a type-I NOPA in a negative uniaxial crystal. The phase-matching and the energy-conservation conditions in the interaction are:

$$\boldsymbol{k}_1^{\mathrm{e}} = \boldsymbol{k}_2^{\mathrm{o}} + \boldsymbol{k}_3^{\mathrm{o}}\,, \tag{1}$$

$$\omega_1 = \omega_2 + \omega_3\,. \tag{2}$$

Here, $\boldsymbol{k}_i$ and ω_i (λ_i) are the wavevector and the frequency (wavelength) of the ith beam, $i = 1, 2$, and 3. Solving (1) and (2), we obtain the following expression of the phase-matching angle (θ), defined as the internal angle made by the extraordinary (e) polarized pump beam with the optical axis of the crystal:

$$\theta = \cos^{-1}\left[\frac{(A^{\mathrm{e}}/Y)^2 - 1}{(A^{\mathrm{e}}/A^{\mathrm{o}})^2 - 1}\right]^{0.5}\,. \tag{3}$$

Here,

$$Y = \sqrt{\left[(k_2^{\mathrm{o}})^2 + (k_3^{\mathrm{o}})^2 + 2k_2^{\mathrm{o}}k_3^{\mathrm{o}}\cos\psi\right]}\,, \tag{4}$$

$$\psi = \alpha + \beta\,, \tag{5}$$

$$\beta = \sin^{-1}\left[(k_2^{\mathrm{o}}/k_3^{\mathrm{o}})\sin\alpha\right]\,, \tag{6}$$

$A^{\mathrm{o}} = 2\pi(n_1^{\mathrm{o}}/\lambda_1)$, $A^{\mathrm{e}} = 2\pi(n_1^{\mathrm{e}}/\lambda_1)$, $k_2^{\mathrm{o}} = |\boldsymbol{k}_2^{\mathrm{o}}| = 2\pi \times (n_2^{\mathrm{o}}/\lambda_2)$, and $k_3^{\mathrm{o}} = |\boldsymbol{k}_3^{\mathrm{o}}| = 2\pi(n_3^{\mathrm{o}}/\lambda_3)$. n_1, n_2, and n_3 are refractive indices of the three interacting radiations with the wavelengths λ_1, λ_2, and λ_3, respectively. The superscripts o and e correspond to the ordinary and extraordinary polarizations, respectively, and α (β) is the noncollinear angle between the pump and the signal (idler) beams. Here k_2^{o} and k_3^{o} are the wave numbers corresponding to the signal and idler beams, whereas Y may be defined as the 'averaged' wave number of the signal and the idler beams parallel to the pump beam. Considering the phase matching along the direction perpendicular to $\boldsymbol{k}_1$, (6) is obtained, and so the approximation made in [20] is not required. In the case of a monochromatic pump, from (3) it is observed that the phase-matching angle θ will be independent (first order) of the signal wavelength or signal frequency (ω_2) if $\partial\theta/\partial\omega_2 = 0$ for any value of ω_2. In (3), all the parameters, except Y, are independent of ω_2. Therefore, the condition may be expressed as follows:

$$\partial Y/\partial\omega_2 = 0\,. \tag{7}$$

It can be shown easily that

$$Y = k_2^{\mathrm{o}}\cos\alpha + k_3^{\mathrm{o}}\cos\beta\,. \tag{8}$$

If the incident angle of the tunable seed pulses have no frequency or equivalently wavelength dependence, the noncollinear angle (α) between the pump and the tunable seed pulses will remain unchanged with the change of the signal frequency, i.e. $\partial\alpha/\partial\omega_2 = 0$. Now from, (7) and (8), we get

$$(\cos\alpha/\nu_{\mathrm{g}2} - \cos\beta/\nu_{\mathrm{g}3}) - k_3^{\mathrm{o}}\sin\beta(\partial\beta/\partial\omega_2) = 0\,, \tag{9}$$

where $\nu_{\mathrm{g}i} = \partial\omega_i/\partial k_i$ is the group velocity of the ith beam ($i = 2$ and 3) with frequency of ω_i. To obtain (9), we have used $\partial\omega_2 = -\partial\omega_3$, since $\omega_1 =$ constant, the pump being monochromatic. From (6) we obtain

$$k_3^{\mathrm{o}}\sin\beta = k_2^{\mathrm{o}}\sin\alpha\,. \tag{10}$$

Differentiating (10) with respect to ω_2, and considering that $\partial\alpha/\partial\omega_2 = 0$ and $\partial\omega_2 = -\partial\omega_3$, it can be shown easily that $\partial\beta/\partial\omega_2 = (\sin\alpha/\nu_{\mathrm{g}2} + \sin\beta/\nu_{\mathrm{g}3})/(k_3^{\mathrm{o}}\cos\beta)$. Substituting the value of $\partial\beta/\partial\omega_2$ in (9), after some algebraic simplification we get $(\cos\psi/\nu_{\mathrm{g}2} - 1/\nu_{\mathrm{g}3})/\cos\beta = 0$, which is equivalent to the following:

$$\nu_{\mathrm{g}2} = \nu_{\mathrm{g}3}\cos\psi\,. \tag{11}$$

The above condition (11) can also be derived [5, 10] by equating the first-order derivative of the phase mismatch with respect to ω_2 to zero, considering a monochromatic pump and $\partial\alpha/\partial\omega_2 = 0$. Earlier, using this method, one author of this paper and his collaborator [5] showed that for a NOPA the effective inverse group-velocity (GV) mismatch between the signal and the idler pulses GVM_{2-3} may be written as follows:

$$\mathrm{GVM}_{2-3} = 1/\nu_{\mathrm{g}2} - 1/(\nu_{\mathrm{g}3}\cos\psi)\,. \tag{12}$$

It is observed from (12) that GVM_{2-3} can be eliminated if the above condition (11) is satisfied and so the condition (11) is called the condition for the GV matching in a NOPA [5]. Therefore, following the earlier method [5, 10] it can be shown only implicitly that the condition (11) is the condition for the insensitivity of the phase-matching angle with respect to the variation of signal frequency or wavelength. However, above we have derived the same condition in a straightforward as well as a simple manner starting from the expression of the phase-matching angle (θ), and it is shown explicitly that, to achieve the insensitivity of the phase-matching angle with respect to the variation of signal frequency or wavelength of the NOPA, the condition (11) is to be satisfied, i.e. the components of the group velocities of the idler beams with a broad spectrum along the signal direction must be equal to those of the corresponding signal beam. In the following we study the optical parametric properties of a relatively recently developed NLO crystal LB4 and find that the above condition (11) is also satisfied if this crystal is employed as a NLO amplifier crystal in a 395-nm-pumped type-I NOPA with a particular value of the noncollinear angle (α).

3 Nonlinear optical properties of LB4 crystal employed in a type-I NOPA for the generation of tunable ultra-fast laser radiation

3.1 *Type-I NOPA phase-matching characteristics*

Figure 2 shows the phase-matching characteristics of the type-I LB4 NOPA pumped by 395 nm, the second harmonic of the Ti : sapphire laser radiation. The phase-matching angles are calculated using (3)–(6), while the refractive indices at the interacting wavelengths are calculated using the Sellmeier dispersion relations of Sugawara et al. [24]. In Fig. 2 the phase-matching tuning curves for both signal and idler branches are shown for the collinear geometry with $\alpha = 0°$ by the solid curve as well as those for the noncollinear geometries with two different values of α of 2.9° and 3.5° by the dashed and the dotted curves, respectively. In Fig. 3 the variation of the propagation directions of the pump, signal, and idler beams with the signal wavelengths with respect to the optical axis of the crystal are shown for $\alpha = 2.9°$; the signal branch has been magnified in a region of wavelengths below 800 nm to show the wavelength insensitivity of the phase

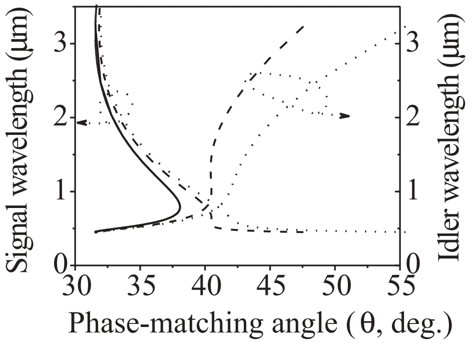

FIGURE 2 Theoretical phase-matching curves (signal and idler branches) of θ in a type-I $Li_2B_4O_7$ (LB4) NOPA, for different internal noncollinear angles (α), pumped by 395-nm radiation and seeded by the white-light continuum. The *solid*, *dashed*, and *dotted curves* are for $\alpha = 0°$, 2.9°, and 3.5° respectively. The region below 0.45 μm of signal wavelength is limited by the idler absorption in the crystal

matching for the amplification of the signal wavelengths below 800 nm. From Fig. 3 it is observed that for $\alpha = 2.9°$ the idler-beam propagation angle (θ_3) is dependent on the signal wavelength and hence the idler wavelength; as the pump is monochromatic its wavelength is constant. On the other hand, the pump (θ) and signal (θ_2) beam-propagation directions are unchanged for the variation of the signal wavelength in certain regions below 800 nm, viz. for the variation of the signal wavelength in the region 525–725 nm θ as well as θ_2 varies within $40.5 \pm 0.1°$. It may be noted that even for a small change of α the phase-matching curve is modified substantially. However, it is found that the phase-matching curves for the signal and pump radiations are almost flat for $\alpha = 2.9°$, and so only this value of α is considered in Fig. 3 as well as in the following calculation of the type-I LB4 NOPA bandwidth.

The wave-vector mismatch in the interaction has been calculated to estimate the phase-matched bandwidth of the process. The wave-vector mismatch (Δk) along the pump direction is given as follows:

$$\Delta k = k_1^{\mathrm{e}}(\theta) - k_2^{\mathrm{o}} \cos\alpha - k_3^{\mathrm{o}} \cos\beta \,. \tag{13}$$

Figure 4 shows the variation of the (normalized) wave-vector mismatch ($\Delta kL/\pi$) with the signal wavelengths (below 800 nm) of a 395-nm-pumped type-I NOPA in different crystals, namely BBO, KABO, CLBO, and LB4. During the numerical calculation of Δk the center wavelength of the signal radiation is considered at 630 nm and the pump-signal noncollinear angles (α) for BBO [1–10], KABO [20], CLBO [20], and LB4 crystals are 3.7°, 3.4°, 3.0°, and 2.9°, respectively; the thickness (L) of all the crystals is taken to be 1 mm. It may be noted that for KABO the optimum value of α obtained with the recent Sellmeier dispersion [19] is 3.4°, instead of 3.3° [20]. From Fig. 4 it is found that in the case of an LB4 crystal with $\Delta kL = \pm\pi$, λ_2 varied over 520 ± 745 nm, which corresponds to the phase-matched bandwidth of over 174 THz, whereas the estimated bandwidths for BBO, KABO, and CLBO crystals are 157, 152, and 164 THz, respectively. The detailed results for CLBO and KABO were presented previously by us [20], whereas those for BBO are well known [1–10]. However, it is observed that the phase-matched bandwidth of a 395-nm-pumped type-I NOPA with the LB4 crystal is larger than that with any of the other three borate-group crystals. In the following we describe the group-velocity matching and some other properties of the considered type-I LB4 NOPA.

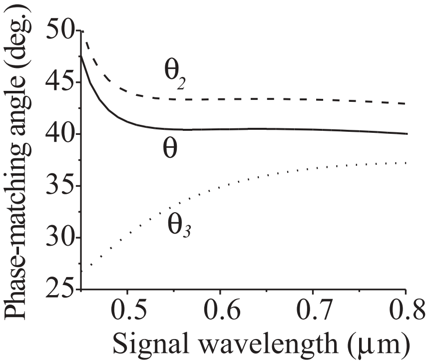

FIGURE 3 Variations of the phase-matching angle corresponding to the pump (θ), signal (θ_2), and idler (θ_3) radiations with the signal wavelength of a 395-nm-pumped type-I LB4 NOPA when the pump-seed noncollinear angle is $\alpha = 2.9°$

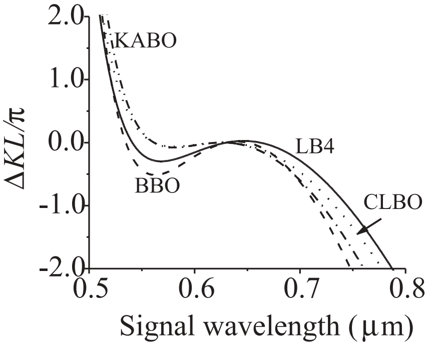

FIGURE 4 The variation of the normalized wave-vector mismatch ($\Delta kL/\pi$) with the signal wavelengths below 800 nm of type-I LB4, BBO, CLBO, and KABO NOPAs pumped by 395-nm radiation. The *solid*, *dashed*, *dotted*, and *dash-dotted curves* are for LB4, BBO, CLBO, and KABO crystals, respectively, and the corresponding values of the (internal) noncollinear angles (α) are 2.9°, 3.7°, 3.0°, and 3.4°, respectively

3.2 *Group-velocity matching and some other properties of a type-I NOPA*

Among others, the inverse GV mismatch between the signal and idler radiations (GVM_{2-3}) of a NOPA is an important parameter as GVM_{2-3} determines predominantly the bandwidth of a NOPA [28]. From (12) it is observed that in the spectral range of normal dispersion ($\nu_{g2} < \nu_{g3}$ for $\lambda_2 < \lambda_3$), GV matching is only allowed in the degeneracy condition for a collinear ($\alpha = 0°$) OPA. On the other hand, by employing noncollinear geometry it is possible to achieve GV matching for a broad range of wavelengths of the signal radiations. For a NOPA, the effective value of GVM_{2-3} is given by (12) and GV mismatch between the pump and the signal beams (GVM_{1-2}) is given by the following equation [5]:

$$\mathrm{GVM}_{1-2} = 1/\nu_{g1} - 1/(\nu_{g2}\cos\alpha) \,. \tag{14}$$

As seen from (14), by employing proper noncollinear geometry GV matching between the pump and the signal is also possible. Di Trapani et al. [4] realized GV matching between the pump and the signal in the case of $\nu_{g1} < \nu_{g2}$, and

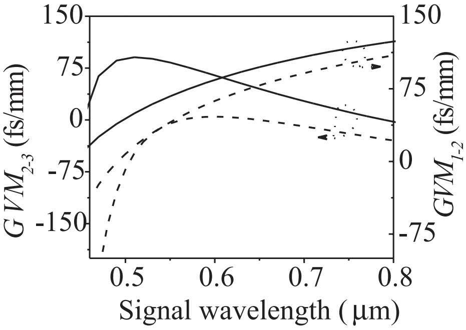

FIGURE 5 Variations of the inverse group-velocity mismatch between the pump–signal (GVM_{1-2}) and signal–idler (GVM_{2-3}) with the signal wavelengths of a 395-nm-pumped type-I LB4 NOPA. The *solid* and *dashed curves* are for $\alpha = 0°$ and 2.9°, respectively

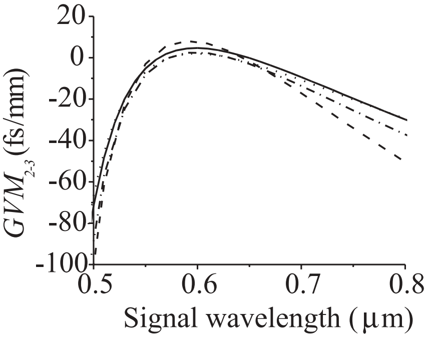

FIGURE 6 The *solid*, *dashed*, *dotted*, and *dash-dotted curves* show the variation of the group-velocity mismatch between the signal and idler radiations (GVM_{2-3}) of the type-I NOPA in LB4, BBO, CLBO, and KABO crystals with the corresponding values of $\alpha = 2.9°$, 3.7°, 3.4°, and 3.0°, respectively

obtained an order of magnitude longer interaction length than that for the collinear configuration. Here Fig. 5 shows the variation of GVM_{2-3} and GVM_{1-2} calculated by using (12) and (14), respectively, for a type-I LB4 NOPA for collinear ($\alpha = 0°$, solid curves) and noncollinear geometries with $\alpha = 2.9°$ (dotted curves). From Fig. 5 it is observed that by employing $\alpha = 2.9°$ it is possible to realize GV matching for a broad range of wavelengths of the signal radiations, viz. the values of GVM_{2-3} remain only within ±20 fs/mm when the values of signal wavelengths varied between 530 and 760 nm, whereas for the collinear configuration GV matching is possible only at a degenerate wavelength. The variations of GVM_{2-3} with the NOPA signal wavelength below 800 nm have also been calculated for other borate-group crystals BBO, KABO, and CLBO, and they are shown together in Fig. 6. From Fig. 6 it is observed that in the signal wavelength regions concerned the maximum value (taking the magnitude) of GVM_{2-3} is smaller for CLBO and LB4 crystals than it is for BBO.

The nonlinear optical coupling coefficient (d_{eff}) of an LB4 type-I NOPA has been compared with those of BBO, CLBO, and KABO crystals. Figure 7a shows d_{eff} calculated for LB4, BBO, CLBO, and KABO type-I NOPAs at a central signal wavelength at 630 nm. For the calculation of d_{eff} the phase-matching angle for each crystal is taken corresponding to the signal wavelength at 630 nm and with $\alpha = 3.7°$, 3.4°, 3.0°, and 2.9° for BBO, KABO, CLBO, and LB4 crystals, respectively. It is observed in Fig. 7a that the LB4 crystal has a rather low value of d_{eff}. However, this disadvantage of a low value of d_{eff} can be overcome by employing tight focusing due to its very high laser damage threshold (LDT) compared to that of the other borate crystals. The LDT of CLBO is ∼ 26 GW/cm^2 for Nd : YAG laser radiation at 1064 nm and for 1.1-ns pulses, which is almost twice as high as that of BBO [29], whereas the same for LB4 crystal [21] is 40 GW/cm^2 for Nd : YAG laser radiation at 1064 nm with a 10-ns pulse width at 10 Hz. These are shown in Fig. 7b. The LDT of KABO is not shown in Fig. 7b since we do not have sufficient data. However, for KABO crystal the only available data for its damage resistance is > 1 GW/cm^2 for repeated pulses and > 15 GW/cm^2 for a single pulse using 1064 nm with 10-ns laser radiation at 10-Hz repetition rate [18]. From Fig. 7b it is seen that the LDT of LB4 crystal is ∼ 3 and 1.5 times that of BBO and CLBO crystals, respectively, and this may be the highest among those of NLO borate crystals [21]. 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 crystal. However, by judicious choice of noncollinear configurations it is possible to overcome the deleterious effect of the walk-off. In the case of BBO crystal ϱ is almost equal to the noncollinear angle (α) required to achieve broadband phase matching and this fortuitous matching is made possible in BBO to achieve large gain as well as large bandwidth. From the results presented in Sect. 3.1 and also in this section, it is found that to achieve broadband amplification of signal wavelengths centered at 630 nm in a 395-nm-pumped type-I LB4 NOPA the required value of α is ∼ 2.9°. The value of ϱ at 395 nm in the case of LB4 crystal is ∼ 2.1°. Therefore, by employing noncollinear geometry it is possible to alleviate the deleterious walk-off effect in a type-I NOPA and achieve the large gain

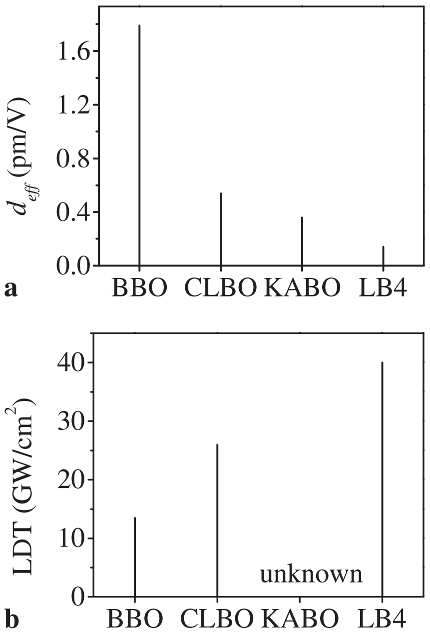

FIGURE 7 **a** The effective nonlinear optical coefficients of BBO, CLBO, KABO, and LB4 crystals for the type-I NOPA. **b** The laser damage threshold (LDT) of BBO, CLBO, and LB4 crystals at Q-switched ns pulsed Nd : YAG laser radiation [21, 29]

as well as bandwidth also in LB4 crystal that was exploited earlier in BBO crystals [1–10]. However, considering that the parametric gain (G) of the NOPA is approximately proportional to $d_{\text{eff}}^2 I_p L^2$, the damage threshold intensity of LB4 is ~ 3 times higher than that of BBO and d_{eff} is $\sim 1/10$th that of BBO; with the highest pump intensity (I_p) the maximum value of G of an LB4 NOPA should be $\sim 3/10^2$ times smaller than that of BBO if the crystal lengths are the same. Taking a thicker crystal length, say 4.5 mm, it may be possible to compensate the low parametric gain. However, the bandwidth of a NOPA with the thicker crystal length is reduced, viz. the estimated bandwidth of an LB4 NOPA is ~ 110 THz with $L = 4.5$ mm and $\alpha = 2.95°$. Taking $\alpha = 2.9°$, which was used in earlier calculations with the 1-mm-thick crystal, the estimated bandwidth is ~ 130 THz for a 4.5-mm-thick LB4 crystal but in this case the phase-matched signal spectrum of a NOPA is intensity modulated; viz. at ~ 565-nm signal wavelength the intensity becomes as low as $\sim 10\%$ of its maximum value.

4 Conclusion

In conclusion, we have presented a straightforward derivation of the condition for broadband phase matching in a type-I NOPA crystal, this being the requirement for the generation of tunable ultra-fast laser radiation from a NOPA pumped by Ti : sapphire second-harmonic radiation and seeded by the white-light continuum. We have also studied the optical parametric properties of a relatively newly developed nonlinear optical borate crystal, LB4. The crystal has the several advantageous properties to be used in different NLO applications, such as (i) wide optical transmission range (160–3500 nm), (ii) easy growth to excellent optical quality with large-size crystals, (iii) high value of the laser damage threshold [21] of $\sim 40\,\text{GW/cm}^2$, which is ~ 3 times that of BBO and ~ 1.5 times that of CLBO, (iv) good mechanical stability, (v) easy treatment of cutting and polishing, (vi) small walk-off angle, and (vii) chemical stability even in water, i.e. nonhygroscopicity. CLBO and BBO crystals are soft and hygroscopic; CLBO is particularly highly hygroscopic and it requires critical maintenance of the environment, such as a temperature as high as 160°C for stable performance. From this study it is also found that it is possible to achieve the broadband phase matching in LB4 crystal and thus it is suited for the generation of the ultra-fast laser radiation by being employed in a 395-nm-pumped type-I NOPA. The phase-matching bandwidth is numerically estimated for 1-mm-thick BBO, KABO, CLBO, and LB4 type-I NOPAs and it is found that the bandwidth with the LB4 crystal is the widest (> 174 THz) among all these crystals, in principle. The only disadvantage of LB4 crystal is that its nonlinear optical coupling coefficient is rather small. However, this disadvantage may be outweighed at least partially by its high value of the laser damage threshold by employing the tight focusing geometries in NLO applications. Therefore, we believe that the results presented here will bring a lot of interest among researchers in this field to develop an ultra-fast laser radiation source using LB4 crystal by employing a Ti : sapphire second-harmonic (i.e. 395 nm) pumped type-I NOPA scheme, considering its several advantageous characteristics for handling and practical applications.

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