Theoretical Investigation of the Generation of Broad Spectrum Second Harmonics in $Pna_{21}$-$Ba_3Mg_3(BO_3)_3F_3$ Crystals

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(Received March 24, 2021 : revised June 9, 2021 : accepted June 21, 2021)

Borate nonlinear optical crystals have been used as frequency conversion devices in many fields due to their unique transparency and nonlinearity from ultraviolet to visible spectral range. In this study, we theoretically and numerically investigate the properties of broadband second harmonic generation (SHG) in the recently reported $Pna_{21}$-$Ba_3Mg_3(BO_3)_3F_3$ (BMBF) crystal. The technique is based on the simultaneous achievement of birefringence phase matching and group velocity matching between interacting waves. We discussed all factors required for broadband SHG in the BMBF in terms of two types of phase matching and group velocity matching conditions, the beam propagation direction and the corresponding effective nonlinearity and spatial walk-off, and the spectral responses. The results show that bandwidths calculated in the broadband SHG scheme are 220.90 nm (for Type I) and 165.85 nm (for Type II) in full-width-half-maximum (FWHM). The central wavelength in each case is 2047.76 nm for Type I and 1828.66 nm for Type II at room temperature. The results were compared with the non-broadband scheme at the telecom C-band.

Keywords : Biaxial birefringence, Borate crystals, Group velocity matching, Second-harmonic generation

OCIS codes : (190.2620) Harmonic generation and mixing; (190.4410) Nonlinear optics, materials; (260.1180) Crystal optics; (260.1440) Birefringence

I. INTRODUCTION

Borate nonlinear optic crystals have been used in visible and ultraviolet (UV) applications due to their unique advantages including wide transparent spectral range on the UV side and high damage threshold [1]. A variety of borate crystals, mainly in three structural categories, have been extensively developed and commercialized: (1) $\beta$-BaB$_2$O$_4$ (BBO) with the basic structural unit of (B$_3$O$_6$)$^{3-}$ group, (2) LiB$_4$O$_7$ (LBO) family with the structural unit of (B$_3$O$_7$)$^{5-}$ group, e.g., CsLiB$_6$O$_{10}$ and CsB$_3$O$_5$, (3) KBe$_2$BO$_3$F$_2$ (KBBF) family with the basic structural unit of (BO$_3$)$^{3-}$ group, e.g., K$_2$Al$_2$B$_2$O$_9$, RhBe$_2$BO$_3$F$_2$, CsBe$_2$BO$_3$F$_2$, Sr$_2$Be$_2$B$_2$O$_7$, BaAl$_2$B$_2$O$_7$, and BaAlBO$_3$F$_2$. In particular, since borate crystals with planar (BO$_3$)$^{3-}$ groups have exhibited large second harmonic generation (SHG) response due to the $\pi$-conjugated molecular orbitals of (BO$_3$)$^{3-}$ units, intensive research is still underway [2–6]. Numerous applications of borate crystals include third, fourth, and fifth harmonic generation of Nd-based laser systems, SHG of Ti:sapphire lasers, deep UV generation below 200 nm, optical parametric oscillation and amplification, and entangled photon-pair generation [1].

Preferred properties of borate crystal as a frequency converter include large laser damage threshold (LDT) for improving efficiency, high thermal conductivity for dissipating heat, and low hygroscopicity and low thermal expansion for practical application. Recently, a new borate crystal Ba$_3$Mg$_3$(BO$_3$)$_3$F$_3$ (BMBF) with two types of...
polymorphs (belonging to space groups Pna2₁ and P6₂m, respectively) has been reported [5]. Of these two, the Pna2₁-BMBF shows large LDT (≈6.2 GW/cm²), deep-UV cutoff edge (≈184 nm), the insolubility in water (i.e., non-hygroscopicity in contrast to BBO), and weak anisotropic thermal expansion. These properties are comparable or superior to those of widely used BBO. As a nonlinear optic frequency converter, SHG was briefly discussed only when the interacting beams propagate along the crystallographic axes [5]. However, details of phase matching (PM), effective nonlinearity, spectral response including bandwidth, and spatial walk-off between the interacting beams, which should be discussed in advance for the SHG, have not yet been reported. Therefore, it is time to investigate the usefulness of Pna2₁-BMBF as another nonlinear optical frequency converter based on borate crystals.

In this paper, we theoretically and numerically investigate the SHG properties of Pna2₁-BMBF. First, we theoretically explain two PM types for the SHG in a Pna2₁-BMBF, namely Type I and Type II, followed by a theoretical summary of group velocity (GV) matching between the interacting waves, the spatial walk-off between them, and the effective nonlinearity. Then, we will discuss a broad spectrum SHG approach in crystals with point symmetry mm2, such as a Pna2₁-BMBF. The technique is based on the simultaneous achievement of birefringence PM and GV matching between interacting waves. In the next section, we explain the simulation results showing that the PM and GV matching conditions can be satisfied simultaneously in a specific range of the direction of the wave vector. This means that the SHG resonance can be tuned on the spectrum by sweeping the wave vector within this range. Then, we discuss the numerical simulation results of effective nonlinearity and spatial walk-off in the broadband SHG, followed by the comparison of spectral responses between the broadband SHG approach and typical SHG case without the GV matching. The results show that the spectral bandwidths are wider by 19.65 times (for Type I) and 10.35 times (for Type II) for the broadband SHG approach than for the typical case.

II. THEORIES

A Pna2₁-BMBF belongs to the orthorhombic mm2 point group. The crystallographic axes of Pna2₁-BMBF are all perpendicular to each other, and the lattice constants are \( a = 0.80740 \text{ nm} \), \( b = 1.53072 \text{ nm} \), and \( c = 0.88218 \text{ nm} \) [5]. When the \( c \)-axis is set to be parallel to the \( x \)-axis, the order of the refractive index (RI) magnitudes is given by \( n_2 < n_3 < n_1 \), showing biaxial birefringence. The Sellmeier equations of Pna2₁-BMBF are found in [5]. The RIs of the two eigen-polarization modes of light traveling inside the Pna2₁-BMBF crystal are expressed as follows by solving the Fresnel equation of the wave normal [7]:

\[
n^{\text{eff}}(j\omega) = \frac{2}{\sqrt{-B_j \pm \sqrt{B_j^2 - 4C_j}}},
\]

The definitions of the parameters in Eq. (1) are as follows:

\[
B_j = -(b_j + c_j)k_j^2 - (a_j + c_j)k_k^2 - (b_j + a_j)k_l^2,
\]

\[
C_j = b_j c_j k_y^2 + a_j c_j k_z^2 + b_j a_j k_z^2,
\]

\[
a_j = n_j(j\omega), \quad b_j = n_j(j\omega), \quad c_j = n_j(j\omega),
\]

where each \( k \) represents the \( x \)-, \( y \)-, and \( z \)-axis components of the wave vector defined in spherical coordinates, i.e., \( \sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta \). Here, \( \theta \) and \( \phi \) are the polar and azimuthal angles, respectively. \( j \) can be either 1 or 2, then \( n(\omega) \) and \( m(2\omega) \) denote the RIs of the fundamental wave and its second harmonics with frequencies \( \omega \) and \( 2\omega \), respectively. \( m \) in Eq. (1) can be either \( h \) or \( l \), representing high or low RI. Here, \( h \) and \( l \) are obtained by taking minus and plus from the \( \pm \) sign of the denominator in Eq. (1), respectively.

Figure 1 illustrates two types of birefringent PM possible in a Pna2₁-BMBF for SHG. In both cases, a pair of input fundamental photons with the same frequency of \( \omega \) generates an SH photon of frequency of \( 2\omega \). For Type I, the polarization state of the SH photon is perpendicular to that of the two fundamental photons, as depicted in Fig. 1(a). On the other hand, in Type II, a pair of fundamental
photons with polarization states perpendicular to each other produces an SH photon [see Fig. 1(b)]. The two types of PM conditions are expressed as follows:

\[ \Delta k = |k^{(1)}(2\omega) - 2k^{(2)}(\omega)| = 0 \text{ (for Type I)} \]
\[ \Delta k = |k^{(2)}(2\omega) - k^{(3)}(\omega) - k^{(1)}(\omega)| = 0 \text{ (for Type II)}. \]

The temporal walk-off (\(\Delta T\)) between the interacting photons due to the difference in GV can be expressed as \(\Delta T\) per unit crystal length as follows:

\[ \Delta T = \frac{\Delta n_g}{L} \tag{7} \]

where \(L\) and \(\Delta n_g\) represent the crystal length and the group index difference between the interacting photons, respectively [8]. Then, GV matching is achieved when \(\Delta n_g = 0\) in Eq. (7), which can be simplified as

\[ n_g^{(1)}(2\omega) = n_g^{(3)}(\omega) \text{ (for Type I)}, \]
\[ \quad 2n_g^{(2)}(2\omega) = n_g^{(3)}(\omega) + n_g^{(1)}(\omega) \text{ (for Type II)}. \]

Each subscript \(g\) in Eqs. (8) and (9) means the group index. Now the broad-spectrum SHG for each case shown in Fig. 1 is defined as Eqs. (5) and (8) (for Type I) or Eqs. (6) and (9) (for Type II) being satisfied simultaneously. Each of these equations is a three-variable function for fundamental wavelength (\(\lambda_r\)), \(\theta\), and \(\phi\). Therefore, by solving a system of Eqs. (5) and (8) (for Type I) or Eqs. (6) and (9) (for Type II) while changing \(\lambda_r\), we can obtain the set of \(\theta\) and \(\phi\), i.e., the direction of beam propagation at the given \(\lambda_r\). Thus, by sweeping the wave vector of the fundamental wave along the direction characterized by the solution set of \(\theta\) and \(\phi\), we can continuously tune the resonant \(\lambda_r\) while maintaining broadband SHG. This means that the spectral position of the SH wave can be selectively determined or tuned within the range of the solution sets. In contrast, for uniaxial crystals (e.g., LiNbO₃ and BBO), the PM and GV matching conditions are both given as two-variable functions of \(\lambda_r\) and \(\theta\), so there can be at most one set of \(\lambda_r\) and \(\theta\) for broadband SHG.

The effective nonlinear optic coefficients of biaxial crystals with point symmetry mm2 are determined by the relationship between the crystallographic axes and the optical axes. For a \(Pna2_1\)-BMBF with the relationships of \((x, y, z) = (c, b, a)\), \(d_{ij}\) is expressed as follows [9]:

\[ d_{ij} = \xi_i d_{15} + \xi_j d_{34} + \xi_{ij} d_{14} + \xi_{ij} d_{32} + \xi_{ij} d_{31} \tag{10} \]

where \(d\)-values in the axial directions are \(d_{15} = d_{31} = 0.09\ pm/V\), \(d_{34} = -0.39\ pm/V\), and \(d_{33} = 0.51\ pm/V\), respectively, when Kleinman symmetry conditions are valid [5]. The subscript \(i\) in Eq. (10) can be either 1 or 2, and represents Type I and Type II, respectively. Then, the \(\xi\)-coefficients for each type are given as follows:

\[ \xi_{i1} = -2A^2E(BGH - CE), \]
\[ \xi_{i2} = -2(BCE - GH)(BCH + CE)(BCH + EG), \]
\[ \xi_{i3} = -A^2H^2(BEG + CH), \]
\[ \xi_{i4} = -(BCH + EG)^2(BEG + CH), \]
\[ \xi_{i5} = -(BCH - CE)^2(BEG + CH), \]
\[ \xi_{i6} = A^2E(BGH - CE) + A^2E(BEG + CH), \]
\[ \xi_{i7} = (BCE - GH)(BCH - CE)(BCH + EG)(BCH + EG)(BEG + CH), \]
\[ \xi_{i8} = A^2E(BEG + CH), \]
\[ \xi_{i9} = (BCE - GH)(BCH + EG)(BEG + CH), \]
\[ \xi_{i10} = (BCH - CE)(BEG + CH)^2, \]

where the angle-dependent parameters of \(A, B, C, G, E, H\) are given by \(\sin \theta\), \(\cos \theta\), \(\sin \phi\), \(\cos \phi\), \(\sin \delta\), and \(\cos \delta\), respectively. The angle \(\delta\) introduced for convenience only is defined as

\[ \tan \delta = \frac{2BCG}{A^2\cot^2 \theta + C^2 - B^2G^2}, \tag{13} \]

where \(V_z\) represents the angle between the z-axis and the optic axis of biaxial birefringence when the relation of \(n_r < n_i < n_s\) is valid, as in the case of the \(Pna2_1\)-BMBF crystal. Since \(V_z\) depends on the Sellmeier equations of the crystal, \(d_{ij}\) is given as a three-variable function for \(\lambda_r\), \(\theta\), and \(\phi\). Thus, \(d_{ij}\) for each type can be obtained by substituting the solution sets of \(\lambda_r\), \(\theta\), and \(\phi\) that satisfy the condition for broadband SHG. The SHG efficiency is proportional to the square of \(d_{ij}\) for a given direction of beam propagation [10].

When a light beam travels inside a biaxial crystal with the point symmetry mm2, the spatial walk-off between the wave vector and the Poynting vector occurs within the crystal [7]. The walk-off angle (\(\rho\)) can be expressed as

\[ \tan \rho^{(m)} = \left[ \frac{k_{ji}}{\left(n^{(m)}(j\omega)\right)^2 - a_j} \right]^2 + \left[ \frac{k_{ji}}{\left(n^{(m)}(j\omega)\right)^2 - b_j} \right]^2 + \left[ \frac{k_{ji}}{\left(n^{(m)}(j\omega)\right)^2 - c_j} \right]^2 \right]^{1/2}, \tag{14} \]
where, all parameters of Eq. (14) are defined in Eqs. (1)–(4). For the Type I case of Eq. (5), an SH photon with a low RI interacts with two high RI fundamental photons with the same polarization state. In this case, the walk-off can be defined as the angle ($w$) between the Poynting vectors of the interacting photons, which is given by [11]

$$w = \cos \rho_1^{(1)} \cos \rho_2^{(1)}.$$  \hspace{1cm} (15)

For the Type II case of Eq. (6), $w$ is defined as the largest angle formed by the Poynting vectors of the interacting beams. Since the low-RI fundamental beam is always placed between the high-RI fundamental beam and the low-RI SH beam, $w$ for Type II can use the same definition as for Type I in Eq. (15). Here, $w$ is also the three-variable function for $\lambda_r$, $\theta$, and $\phi$, which can be obtained by substituting the solution sets of $\lambda_r$, $\theta$, and $\phi$ that satisfy the condition for broad-spectrum SHG in each case. Note that even with the same definition of $w$, the calculation result will be different for each type because the solution sets of $\lambda_r$, $\theta$, and $\phi$ depend on the interaction type. The maximum deviation between the interacting beams after passing through a crystal of length $L$ is expressed as

$$\Delta = L \tan w.$$  \hspace{1cm} (16)

### III. RESULTS AND DISCUSSION

Figure 2(a) shows the PM properties of a Pna2$_1$-BMBF crystal for SHG. The red and the blue surfaces represent Type I and Type II PM calculated using Eqs. (5) and (6), respectively. The vertical axis of $\lambda_r$ covers the entire transparent range of the crystal, and the base of the coordinates is the plane formed by the angles $\theta$ and $\phi$ representing the direction of the wave vector. First, we discuss the non-critical PM (NCPM) properties where GV matching is not considered. Here, the NCPM refers to PM when the interacting beams propagate along one of the crystallographic axes. In this case, no spatial walk-off occurs between the interacting beams, leading to a longer length of nonlinear optic interaction within the crystal and thus higher SHG ef-

![FIG. 2. The PM characteristics and the corresponding effective nonlinearity properties: (a) PM properties of a Pna2$_1$-BMBF for SHG; PM curves (the solid black lines) and density plots of $d_{\text{eff}}$ for (b) Type I and (c) Type II.](image)

**TABLE 1.** Non-critical phase matching (NCPM) properties of a Pna2$_1$-BMBF in SHG

<table>
<thead>
<tr>
<th>$k$-direction</th>
<th>$\theta$ [°]</th>
<th>$\phi$ [°]</th>
<th>$\lambda_r$ [nm]</th>
<th>$d_{\text{eff}}$</th>
<th>Polarization</th>
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<tbody>
<tr>
<td><strong>Type I</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>z-axis</td>
<td>0</td>
<td>0</td>
<td>641.91</td>
<td>0</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0</td>
<td>90</td>
<td>626.70</td>
<td>$d_{31}$</td>
<td>$z + z \rightarrow x$</td>
</tr>
<tr>
<td>y-axis</td>
<td>90</td>
<td>90</td>
<td>927.11</td>
<td>$d_{34}$</td>
<td>$x + y \rightarrow y$</td>
</tr>
<tr>
<td><strong>Type II</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>z-axis</td>
<td>0</td>
<td>0</td>
<td>927.11</td>
<td>0</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0</td>
<td>90</td>
<td>3450.83</td>
<td>$d_{34}$</td>
<td>$x + y \rightarrow y$</td>
</tr>
<tr>
<td>y-axis</td>
<td>90</td>
<td>90</td>
<td>900.45</td>
<td>0</td>
<td>-</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>3125.82</td>
<td>0</td>
<td>-</td>
</tr>
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ficiency. As can be seen in Fig. 2(a), the NCPM is possible for \((\theta, \phi) = (0^\circ, 0^\circ), (0^\circ, 90^\circ), (90^\circ, 90^\circ)\). Here, the interacting beams propagate along the \(z\)-axis in the first two cases, and along the \(y\)-axis in the third case. The NCPM properties of a \(Pna_2\text{I}_2\)-BMBF in SHG are summarized in Table 1. For Type I, the \(\lambda_F\) values for NCPM are 626.70 nm and 641.91 nm, where non-zero \(d_{\text{eff}}\) coefficients used for the interactions are \(d_{31}\) and \(d_{32}\), respectively. The polarization direction of the fundamental wave is parallel to the \(z\)-axis for \(d_{31}\) and the \(y\)-axis for \(d_{32}\). On the other hand, the polarization of the SH wave is towards the \(x\)-axis in both cases. In the case of Type II, NCPM is achieved only when the interacting beams, the corresponding beam deviation (\(\Delta\)) calculated at the optical communication wavelength of 1550 nm is shown as the density plot in each figure, which is calculated using Eqs. (10)-(12). Here, \(d_{\text{eff}}\) increases in the order of blue, green, yellow, orange, and red zones. For Type I, the maximum \(d_{\text{eff}}\)-value for SHG is 0.217 pm/V, where the corresponding \(k\)-direction is \((\theta, \phi) = (59.3^\circ, 9.8^\circ)\). For Type II, the maximum \(d_{\text{eff}}\) and the corresponding \(k\)-direction are 0.291 pm/V and \((\theta, \phi) = (41.7^\circ, 0^\circ)\), respectively. The walk-off angles (\(w\)) calculated using Eq. (15) are 1.43° (for Type I) and 1.23° (for Type II). The corresponding beam deviations per unit millimeter crystal length calculated using Eq. (16) are 25.0 \(\mu\text{m/mm}\) (for Type I) and 21.5 \(\mu\text{m/mm}\) (for Type II), which can be sufficiently overcome by using a larger sized pump beam in thick crystals. The \(k\)-directions for PM, \(d_{\text{eff}}\)-values, the walk-off angles (\(w\)), and the corresponding beam deviation (\(\Delta\)) calculated at 1550 nm are listed in Table 2. Figure 3 shows the spectra of fundamental waves that are acceptable for SHG – Fig. 3(a) for Type I and Fig. 3(b) for Type II. The vertical axis of each graph represents the transmittance (or normalized intensity). The nonlinear optic interaction length of 10 mm in the crystal was used in the calculation. As shown in Fig. 3, both spectra show typical sinc-function shapes. The calculated bandwidths are 11.24 nm (for Type I) and 16.03 nm (for Type II) in full-width-half maximum (FWHM). The results will be compared in the next paragraph with the case of the broadband SHG where the PM and the GV matching are considered simultaneously.

Now we discuss the SHG properties at the optical communication wavelength of 1550 nm as a common example where GV matching is not considered. The reason that the communication C-band, not the UV region, was chosen as a comparison group is that the shortest values of \(\lambda_F\) that satisfies the BPM for SHG are 626.70 nm and 641.91 nm for Type I, and 927.11 nm for Type II, and thus are not in the UV region. This can be clearly seen from the NCPM conditions in Fig. 2(a) and Table 1. Therefore, the C-band, where BPM is possible for both types while GV matching is not possible, was chosen as a comparison group. Broadband SHG based on simultaneous quasi-phase matching and GV matching at the C-band in MgO-doped periodically poled lithium niobate crystals have been reported elsewhere [12, 13].

The solid black lines in Figs. 2(b) and 2(c) represent the PM curves for Type I and Type II, respectively. The magnitude of \(d_{\text{eff}}\)-value in the given \(k\)-direction (i.e., \(\theta\) and \(\phi\)) is shown as the density plot in each figure, which is calculated using Eqs. (10)-(12). Here, \(d_{\text{eff}}\) increases in the order of blue, green, yellow, orange, and red zones. For Type I, the maximum \(d_{\text{eff}}\)-value for SHG is 0.217 pm/V, where the corresponding \(k\)-direction is \((\theta, \phi) = (59.3^\circ, 9.8^\circ)\). For Type II, the maximum \(d_{\text{eff}}\) and the corresponding \(k\)-direction are 0.291 pm/V and \((\theta, \phi) = (41.7^\circ, 0^\circ)\), respectively. The walk-off angles (\(w\)) calculated using Eq. (15) are 1.43° (for Type I) and 1.23° (for Type II). The corresponding beam deviations per unit millimeter crystal length calculated using Eq. (16) are 25.0 \(\mu\text{m/mm}\) (for Type I) and 21.5 \(\mu\text{m/mm}\) (for Type II), which can be sufficiently overcome by using a larger sized pump beam in thick crystals. The \(k\)-directions for PM, \(d_{\text{eff}}\)-values, the walk-off angles (\(w\)), and the corresponding beam deviation (\(\Delta\)) calculated at 1550 nm are listed in Table 2. Figure 3 shows the spectra of fundamental waves that are acceptable for SHG – Fig. 3(a) for Type I and Fig. 3(b) for Type II. The vertical axis of each graph represents the transmittance (or normalized intensity). The nonlinear optic interaction length of 10 mm in the crystal was used in the calculation. As shown in Fig. 3, both spectra show typical sinc-function shapes. The calculated bandwidths are 11.24 nm (for Type I) and 16.03 nm (for Type II) in full-width-half maximum (FWHM). The results will be compared in the next paragraph with the case of the broadband SHG where the PM and the GV matching are considered simultaneously.

Figure 4 shows the PM and GV matching properties of the \(Pna_2\text{I}_2\)-BMBF– Fig. 4(a) for Type I and Fig. 4(b) for Type II. In each graph, the PM and GV surfaces represent PM and GV matching numerically calculated using Eqs. (5) and (8) (for Type I) or Eqs. (6) and (9) (for Type II).

**TABLE 2.** The \(k\)-directions for PM, the effective nonlinear optic coefficients (\(d_{\text{eff}}\)), the spatial walk-off angles (\(w\)) between the interacting beams, the corresponding beam deviation (\(\Delta\)) calculated at the optical communication wavelength of 1550 nm

|       | \(\theta\) [°] | \(\phi\) [°] | \(|d_{\text{eff}}|\) [pm/V] | \(w\) [°] | \(\Delta\) [\(\mu\text{m/mm}\)] |
|-------|----------------|----------------|-----------------------------|-----------|-----------------------------|
| Type I| 59.3           | 9.8            | 0.217                       | 1.4       | 25.0                        |
| Type II| 41.7           | 0              | 0.291                       | 1.2       | 21.5                        |

**FIG. 3.** The spectra of fundamental waves that are acceptable for SHG: (a) Type I and (b) Type II.
The intersection line of the two surfaces spans a specific range of $\lambda_r$, corresponding to the range of $k_r$-direction (i.e., $\phi$ and $\theta$) that simultaneously satisfy PM and GV matching. The ranges of $\lambda_r$ resonance for broad spectrum SHG are 1752.30 nm–2105.53 nm (for Type I) and 1767.84 nm–1828.65 nm (for Type II), which correspond to the changes in $\lambda_r$ resonance ($\equiv \delta\lambda_r$) of 353.23 nm and 60.81 nm, respectively. These spectral ranges include absorption lines of gas molecules such as methane (CH$_4$), nitric oxide (NO), water vapor (H$_2$O), hydrogen chloride (HCl) and ammonium (NH$_3$). The presence of such absorption lines has applications in many fields, including emission control of exhaust and flue gases, leakage control in gas pipelines, monitoring of breathing gases, monitoring of climate processes, optimization of internal combustion engines, quality control of natural gas pipelines, etc. The $k_r$-directions (i.e., $\phi$ and $\theta$), the ranges of $\lambda_r$ resonance, and $\delta\lambda_r$ are summarized in Table 3.

<table>
<thead>
<tr>
<th></th>
<th>$\phi$ [$^\circ$]</th>
<th>$\theta$ [$^\circ$]</th>
<th>$\lambda_r$ [nm]</th>
<th>$\delta\lambda_r$ [nm]</th>
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<tr>
<td>Type I</td>
<td>0–27.7</td>
<td>59.7–90</td>
<td>1752.30–2105.53</td>
<td>353.23</td>
</tr>
<tr>
<td>Type II</td>
<td>0–43.7</td>
<td>43.2–90</td>
<td>1767.84–1828.65</td>
<td>60.81</td>
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The magnitude of the nonlinear optic coefficient ($d_{eff}$) is determined by the $k_r$-direction as described in the paragraphs with Eqs. (10)-(12). Since the efficiency of SHG is proportional to the square of $d_{eff}$, it is important to estimate the $d_{eff}$-values for the $k_r$-direction for broadband SHG. The $d_{eff}$-values were calculated numerically using Eqs. (10)-(12), the results are plotted in Fig. 5(a) (for Type I) and Fig. 5(b) (for Type II) as functions of $\lambda_r$. Here, the $\lambda_r$-ranges on the horizontal axes correspond to the intersection lines shown in Fig. 4. The largest values of $d_{eff}$ are 0.207 pm/V at 2047.76 nm for Type I and 0.284 pm/V at 1828.66 nm for Type II. The $k_r$-directions (i.e., $\phi$, $\theta$), the ranges of $\lambda_r$ resonance, and $\delta\lambda_r$ are summarized in Table 3.
transmission of optical pulse signals containing information, and nonlinear optical signal processing [15].

**IV. CONCLUSION**

We have theoretically and numerically investigated the broadband SHG in a $Pna_{2}1$-BMBF. In the broadband SHG approach and a typical case without GV matching, PM and GV matching properties, effective nonlinearities, spatial walk-offs, spectral responses were first described, and the results were compared with each other. The ranges of $\lambda_F$ resonance for broadband SHG span 1752.30 nm–2105.53 nm (for Type I) and 1767.84 nm–1828.65 nm (for Type II). The calculated bandwidths are 220.90 nm (for Type I) and 165.85 nm (for Type II) in FWHM. These results are 19.65 times (for Type I) and 10.35 times (for Type II) broader than the common cases without the GV matching. Our studies have shown that BMBF has the following advantages over other borate crystals such as BBO and BiBO. First, in BMBF, the resonance of broadband SHG can be tuned over a wide spectral range of interest, whereas BBO is a uniaxial crystal, exhibiting only one resonance at most. Second, the BMBF is advantageous for packaging due to its low hygroscopicity and high LDT, whereas the BBO is highly hygroscopic and requires strict control of experimental conditions. In the case of BiBO, since the point symmetry is 2 (monoclinic), the crystallographic axes are not perpendicular to each other and are deviated from the optical axes. Thus, non-collinear PM is required, resulting in the large spatial walk-off between the interacting beams. The walk-off angle calculated for BBO and BiBO are 3.5° and 2.3°, respectively, which are larger than 1.3°–1.5° in BMBF for broadband SHG [12]. Therefore, for BMBF, longer nonlinear interaction lengths within the crystal can be used for higher SHG efficiency. Our study is expected to provide versatility as a novel nonlinear optic material platform for applications of borate crystals.

**FIG. 5.** The nonlinear optic coefficient ($d_{eff}$) and the spatial walk-off angles ($\omega$) between the interacting waves plotted as functions of $\lambda_F$ for the broad spectrum SHG: (a) Type I and (b) Type II.

**FIG. 6.** The spectra of fundamental waves that are acceptable for broad spectrum SHG: (a) Type I and (b) Type II.
ACKNOWLEDGMENT

This study was supported by a grant from National Research Foundation of Korea (NRF-2019R1F1A1063937 and the Korea Institute of Science and Technology (KIST) (2E29580-19-147); Institute for Information & communications Technology Promotion (IITP) grant funded by the Korea government (MSIT) (No. 2020-0-00947).

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