Thermal expansion, normalized thermo-optic coefficients, and condition for second harmonic generation of a Nd:YAG laser with wide temperature bandwidth in RbTiOPO₄

Jacques Mangin,1,* Gabriel Mennerat,2 and Philippe Villeval3

1Institut Carnot de Bourgogne, UMR 5206 CNRS-Université de Bourgogne, 9 Avenue A. Savary, BP 47870, F-21078 Dijon Cedex, France
2Commissariat à l’Énergie Atomique/Centre Scientifique et Technique d’Aquitaine, 15 Avenue des Sablères, 33114 Le Barp, France
3Cristal Laser S. A., Parc d’Activités du Breuil, 32 rue Robert Schumann, 54850 Messuin, France
*Corresponding author: jacques.mangin@u-bourgogne.fr

Received October 28, 2010; revised January 18, 2011; accepted January 20, 2011; posted January 20, 2011 [Doc. ID 137320]; published March 23, 2011

Interferometric determination of thermal expansion and of normalized thermo-optic coefficients of RbTiOPO₄ at four laser wavelengths are performed as a function of temperature. A suitable vectorial formalism applied to obtained data allows the establishment of the temperature dependence of refractive indices, and subsequent theoretical analysis enables one to predict that an extremum in the evolution of the phase-matching direction in the (X, Y) plane should occur near 100°C for type II second harmonic generation of Nd:YAG lasers, with a temperature bandwidth that can be as large as 117°C for a crystal of 10 mm in length. Such unusual behavior is observed experimentally by recording the conversion efficiency from 20°C up to 220°C for various propagation angles of light in the (X, Y) plane. Slight quadratic temperature dependence of the effective nonlinear coefficient is also observed. © 2011 Optical Society of America

OCIS codes: 260.3090, 160.4330, 120.6810, 190.4400, 190.2620.

1. INTRODUCTION

Like the well-known KTiOPO₄ (KTP) crystal, the isomorphic RbTiOPO₄ (RTP) is used in a variety of nonlinear optical (NLO) devices, including efficient frequency doubling of high-power Nd:YAG or Nd:YVO₄ radiation to the green for a broad range of medical, industrial, and scientific applications. However, these materials may suffer from a deleterious photochromic effect called "gray tracking," whereby the generated intense green beams alter both refractive indices and transparency [1,2]. This affects the conversion efficiency after a time, depending on crystal dielectric properties, temperature, and illumination conditions. To overcome this drawback, frequency doublers are often heated to 80°C–100°C in demanding systems. Phase-matching conditions and tolerances for frequency doubling are established from dispersion laws of refractive indices, which are generally determined at room temperature. Since refractive indices are temperature dependent, new phase-matching conditions must be well estimated to achieve the best affordable operation at elevated temperatures. This observation holds even more for periodically poled RTP structures, designed to generate tunable radiations and which require, in addition, an accurate knowledge of the dispersion coefficients that govern changes in grating periods with temperature [3,4]. Besides its attractive properties in electro-optics, it has been shown recently that, despite a slightly lower value of the nonlinear effective coefficient $d_{31}$, RTP competes quite well with KTP for the second harmonic generation (SHG) of Nd:YAG lasers at high power levels, due to conjugate higher damage threshold, less sensitivity to photochromic effects, lower walk-off angle, and easiest reliable mastering conditions of domain reversal for manufacturing periodically poled structures [5]. The purpose of this work is to give complete and accurate data on the temperature dependence of both dilatation and of refractive indices of RTP over its whole transparency window and which are actually known from designers to be crucial parameters needed for proper implementation in laser schemes. In addition we give a comprehensive analysis of the evolution of the phase matching angle $\varphi$ for type II SHG of Nd:YAG lasers in the (X, Y) plane of RTP, which is confirmed experimentally by measurements of conversion efficiency performed from 20°C up to 220°C for various propagation angles of light in the (X, Y) plane. The conditions that are to be fulfilled to obtain large temperature bandwidth are also examined.

2. TEMPERATURE-DEPENDENT OPTICAL DISPERSION OF RTP

A. Methodology

We have detailed in previous works the general methodology developed to get a rigorous formulation of the temperature-dependent dispersion equation of optical materials over their whole range of transparency. It is based on absolute interferometric measurements of changes in geometrical length and optical thickness of small parallelepiped-shaped samples that are submitted to linear ramps of temperature over chosen intervals; induced fringe shifts are continuously recorded as a function of temperature [6–8]. If $L$ is the geometrical length of the sample and $n$ its refractive index, the linear thermal

0740-3224/11/040873-098815.00 © 2011 Optical Society of America
expansion coefficient \( \alpha(T) = (1/L) \cdot (dL/dT) \) is obtained by using a modified Mach–Zehnder arrangement acting as an optical gauge, while changes in optical thickness \( \gamma(T) = (1/nL) \cdot (dn/dT) \) of the same sample are given by Fabry–Perot temperature scanning interferometry performed at discrete laser wavelengths \( \lambda_i \) [9]. Both experiments may be conducted over a temperature interval extending from \(-150^\circ \text{C}\) up to \(+300^\circ \text{C}\) and in vacuum enclosures. The normalized thermo-optic coefficient (NTOC) \( \beta \), which we defined as \( \beta = (1/n) \cdot (dn/dT) \), is obtained from the straightforward relationship \( \beta = \gamma - \alpha \).

Now, from the theoretical point of view [8], the dispersion of the refractive index \( n \) of an isotropic optical material with wavelength \( \lambda \) is usually described by the Sellmeier formula, which can be written as

\[
n^2(\lambda, T) = A(T) + \sum_{i=1}^{l} \frac{B_i(T)}{\lambda_i^2 - A_i^2(T)}.
\]

(1)

In the case of anisotropic biaxial RRT, Eq. (1) holds for each one of the three principal refractive indices \( n_x, n_y, \) and \( n_z \) and relevant additional subscripts \( x, y, \) and \( z \) will be appropriately introduced in Subsection 2.8.

The dependence with temperature \( T \) in Eq. (1) is expressed through parameters \( A(T), B_i(T), \) and \( \lambda_i(T); i \) is the number of oscillators \( p \) resonant at wavelengths \( \lambda_i \) that are considered to account for UV as well as IR cutting edges. From Eq. (1), we obtain an analytical expression of the NTOC as function of both wavelength and temperature:

\[
\beta(\lambda, T) = \frac{1}{2 \pi n^2(\lambda, T_0)} \frac{dA}{dT} + \sum_{p=1}^{l} \frac{B_p(T)}{\lambda_i^2 - A_i^2(T)} \frac{dA}{dT} + \sum_{p=1}^{l} \frac{2 B_p(T) \lambda_i (A_i(T)) dA}{\lambda_i^2 - A_i^2(T)^2}.
\]

(2)

On the other hand, in analogy with the linear thermal expansion coefficient, the \( \beta \) data may always be accurately fitted in with a power series of temperature expressed in Celsius degrees:

\[
\beta(\lambda, T) = c_0(\lambda) + c_1(\lambda)T + \ldots + c_m(\lambda)T^m = \sum_{p=0}^{m} c_p(\lambda)T^p.
\]

(3)

The dispersion coefficients \( c_p(\lambda) \) in Eq. (3) must be consistent with Eq. (2) and are thus given by

\[
c_p(\lambda) = \frac{1}{2 \pi n^2(\lambda, T_0)} \left[ X_1 + \sum_{p=1}^{l} \frac{1}{[\lambda_i^2 - A_i^2(T)]} X_p \right] + \sum_{p=1}^{l} \frac{2 B_p(T) \lambda_i (A_i(T)) dA}{\lambda_i^2 - A_i^2(T)^2}.
\]

(4)

The unknown parameters \( X_1 = dA/dT, X_p = dB_p/dT \), and \( X_p = dB_p/dT \) are determined from a simple vectorial formalism applied to the NTOCs that are measured at \( 2^l + 1 \) laser wavelengths \( \lambda_i \) and suitably chosen in the transparency window of the material. Hence, knowing the \( c_p \), the temperature-dependent dispersion equations of the refractive index over the whole useful spectral range is obtained by integrating formula \( \beta = (1/n) \cdot (dn/dT) \) and Eq. (3) and taking into account the dispersion equation known at room temperature \( T_0 = 20^\circ \text{C} \); this gives

\[
n(\lambda, T) = n(\lambda, T_0) \exp \left[ \sum_{p=0}^{m} \frac{c_p(\lambda)}{p+1} (T^{p+1} - T_0^{p+1}) \right].
\]

(5)

B. Experimentation

1. Thermal Expansion and NTOCs

Orthorhombic RTO belongs to the mm2 point group symmetry; it is optically biaxial, and three parallel flat-faced-shaped samples cut along the \( X, Y, \) and \( Z \) axes of the dielectric frame were used to determine all relevant coefficients. They were cut in a flux-grown boule manufactured by Cristal Laser S. A. in a crystal region of high optical homogeneity. The \( X, Y, \) and \( Z \) optical apertures were 5 mm x 5 mm for a geometrical length of 6.3 mm between the parallel optical end faces. Interferometric measurements of the principal thermal expansion coefficients of RTO have been obtained from \(-30^\circ \text{C} \) to \(+130^\circ \text{C} \) by recording continuously the fringe shift with temperature and by using a frequency-stabilized He–Ne laser [10]. The NTOCs were determined over the same temperature interval from measurements of normalized coefficients of changes in optical thickness performed at four CW laser wavelengths and with appropriate polarization direction of the light with respect to the \( X, Y, \) and \( Z \) axes. Monomode CW lasers were used: an Argon ion tuned at 0.4578 µm, two He–Ne emitting respectively at 0.6328 and 3.39 µm, and a Nd:YAG at 1.0642 µm. The experimental results are summarized in Table 1; we notice linear temperature dependence for the dilatation coefficients, with \( \alpha_z < 0 \), while a quadratic one is observed for the NTOCs. Because it is of importance for quasi-phase-matching (QPM) frequency conversion in periodically poled RTO (ppRTO) structures, the dilatation coefficient \( \alpha_z \) is displayed in Fig. 1 for comparison with previous values [3,4].

Measurement uncertainties on thermal expansion and NTOCs were 5.1 \times 10^{-9} K^{-1} and 3.1 \times 10^{-9} K^{-1}, respectively. Since three crystals were used with propagation along the three principal axes along with two possible orientations of the light.

<table>
<thead>
<tr>
<th>Table 1. Polynomial Fits of the Principal Linear Thermal Expansion and NTOCs of RbTiOPO_4 (resp. ( \beta_0 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coefficients</td>
</tr>
<tr>
<td>-------------</td>
</tr>
<tr>
<td>( \alpha_x )</td>
</tr>
<tr>
<td>( \alpha_y )</td>
</tr>
<tr>
<td>( \alpha_z )</td>
</tr>
<tr>
<td>( \beta_0 ) (°µm)</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>( \beta_0 ) (°µm)</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>( \beta_0 ) (°µm)</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>

*°T is the temperature expressed in degrees Celsius. The deviation of experimental data from the fits does not exceed 2 \times 10^{-4} K^{-1} for all coefficients.
polarization in each case, we were able to determine two values for each NTOC from measurements of changes in optical lengths using two crystal orientations. All pairs of $\beta_{01}$, $\beta_{02}$, and $\beta_{03}$ were found to be very consistent, within a difference that did not exceed the nominal uncertainty of $3 \times 10^{-6}$ K$^{-1}$ of each determination. This observation confirmed the high optical homogeneity of the crystal region chosen as well as the reliability of the measured thermal expansion coefficients.

2. Temperature-Dependent Sellmeier Equations

Following the methodology described in Subsection 2A, the determination of the temperature-dependent Sellmeier equations of RTP requires at first the knowledge of the spectral dispersion at a fixed temperature $T_0$. A two-pole equation has been proposed recently at $T_0 = 20^\circ$C that seems well adapted to fit various NLO processes with good accuracy [11] and that we write as

$$n_i^2 = A_i + \sum_{p=1}^{3} \frac{B_{i,p}}{\lambda^2 - \lambda_{p}^2},$$

(6)

The parameters $A_i$, $B_{i,p}$, and $\lambda_{p}$ correspond to each one of the principal axes $i = X, Y, Z$ of the dielectric frame of the crystal are summarized in Table 2; $p = 1, 2$ stands for the poles that are considered, and we have set for convenience $\lambda_{1}^2 = C_1$ and $\lambda_{2}^2 = E_1$.

Following our analysis, a complete formulation of the temperature-dependent dispersion laws $n_i(A, T)$ would require the knowledge of NTOCs at five discrete laser wavelengths. In fact, the IR poles (parameters $E_1$) correspond to lattice absorptions located beyond $\lambda_0 = 11.5 \mu m$ for the three principal directions of polarization, far from the IR cutting edge of spectral transparency. Likewise in the case of HgGa$_2$S$_4$ [8], we will assume that $dE_1/dT = 0$ and keep all other parameters as temperature dependent. Under this assumption, measurements of NTOCs at only four discrete wavelengths are necessary, and Eq. (2) reduces to

$$n_i^2 = A_i + \sum_{p=1}^{3} \frac{B_{i,p}}{\lambda^2 - \lambda_{p}^2} + \frac{C_{i}(T)}{\lambda^2 - C_{i}(T)},$$

(7)

with the dispersion coefficients

$$c_i^0(T) = \frac{1}{2\pi^2 \lambda_i^2(T)} \left[ -58.4301139 + \frac{0.07288812}{(\lambda^2 - 0.047500)} 
+ \frac{0.0235581}{(\lambda^2 - 0.047500)^2} - \frac{838.255985}{(\lambda^2 - 130.76841)} \right],$$

$$c_i^1(T) = \frac{1}{2\pi^2 \lambda_i^2(T)} \left[ 0.8203647 - \frac{0.0110634}{(\lambda^2 - 0.047500)} 
+ \frac{0.0035983}{(\lambda^2 - 0.047500)^2} + \frac{81.4462068}{(\lambda^2 - 130.76841)} \right],$$

$$c_i^2(T) = \frac{1}{2\pi^2 \lambda_i^2(T)} \left[ -0.0012439 - \frac{0.0003989}{(\lambda^2 - 0.047500)} 
+ \frac{0.0003600}{(\lambda^2 - 0.047500)^2} \right],$$

(8)

Applying the vectorial formalism to experimental beta coefficients given in Table 1, we have found the following expressions for the wavelength and temperature-dependent NTOCs:

$$10^6 \times \beta_{i}(A, T) = c_i^0(A) + c_i^1(A)T + c_i^2(A)T^2,$$

(9)

The parameters $A_i$, $B_{i,p}$, and $\lambda_{p}$ corresponding to each one of the principal axes $i = X, Y, Z$ of the dielectric frame of the crystal are summarized in Table 2; $p = 1, 2$ stands for the poles that are considered, and we have set for convenience $\lambda_{1}^2 = C_1$ and $\lambda_{2}^2 = E_1$.
These relationships enable then to determine the principal refractive indices $n_x$, $n_y$, and $n_z$ at any temperature $T$ and wavelength $\lambda$ over the whole transparency window of the material by using Eqs. (5) and (8). The room temperature dispersion of the NTOCs of RbTiOPO$_4$ is drawn from 0.3 $\mu$m to 4 $\mu$m in Fig. 2 along with the results of previous works given for comparison. Identical dispersion curves of NTOCs were obtained by starting from Sellmeier equations reported earlier [12,13] but, of course, with different analytical expressions of the $c_6$. No such attempt was made by using the formulas of [14] because they do not satisfy to Eq. (1) with integer exponents according to the theoretical development detailed elsewhere [15]. The main observation is a significant discrepancy between our results and previously proposed ones, especially in the IR spectral region.

3. THERMAL BEHAVIOR OF THE SHG IN THE $(X,Y)$ PLANE OF RTP

A. Experimentation

Measurements of doubling efficiency were performed on two parallelepipedic-shaped RTP crystals cut at $\theta = 90^\circ$ and $\varphi = \varphi_0 = 55.82^\circ \pm 0.02^\circ$. $\varphi$ is the internal propagation angle of the fundamental beam referred to the principal X axis and $\theta$ its azimuth in the $(X,Y,Z)$ frame. From Eq. (6) and Table 2, this orientation corresponds to type II phase-matched SHG $1.0642 \mu m \rightarrow 0.5321 \mu m$ in the $(X,Y)$ plane at $T_0 = 20^\circ C$. Dimensions of optical apertures were 3 mm x 3 mm and 4 mm x 4 mm for lengths $L = 5.2$ mm and 15 mm, respectively. Two different geometrical lengths were used to better enhance later the difficulty of defining a thermal acceptance for the SHG process. They were illuminated at low power level by a pump beam of a Q-switched injection-seeded Nd:YAG laser, emitting about 5 mJ per pulse of 15 ns duration (FWHM) at a repetition rate of 10 Hz. A tight spatial filtering produced a quasi-Gaussian beam in the near field, with a $1/e^2$ radius $w_0 = 0.85$ mm at the waist. The crystals were inserted in a temperature-regulated oven that was computer controlled and stepped from 20°C to 230°C up and down, with an accuracy of 0.1°C. Step height (10°C) and soaking time were optimized to avoid any temperature hysteresis in optical measurements between upwards and downwards ramps. The oven itself was mounted on a rotation stage to adjust the $\varphi$ angle in the $(X,Y)$ plane with a reading accuracy of 1 arc min. The conversion efficiency was recorded at numerous temperatures and for various $\varphi$ angles chosen in close vicinity of the initial value $\varphi_0 = 55.82^\circ$. Care was taken to stay inside the respective angular acceptance of each crystal, corresponding to $L \Delta \varphi = 0.72^\circ$ cm at full width. The results obtained for the crystal lengths of 5.2 mm and 15 mm are plotted in Figs. 3 and 4, respectively. In both cases and for several incidence angles, it appears that two maxima of efficiency may be observed respectively at “low” and “high” temperature. This is particularly evidenced in Fig. 4 in the case of the longest sample, for which the two peaks are clearly separated. On Fig. 3, for a smaller length, both maxima seem to collapse in a single one on going from $\varphi = 55.45^\circ$ up to $\varphi = 56.70^\circ$, with a peak of efficiency located around 100°C and for a propagation angle close to 66.30°. Solid curves in Figs. 3 and 4 are derived from the theoretical analysis given in Subsection 3.B. The dashed curve in Fig. 3 features the best condition of SHG that can be expected with the crystal of 5.2 mm in length and that occurs at $T_m = 95^\circ$C and $\varphi_m = 56.33^\circ$. In addition, the curve in open black circles in Fig. 4 exhibits also for illustration the optimized configuration calculated for crystals of conventional length $L = 10$ mm.

<table>
<thead>
<tr>
<th>Index</th>
<th>$A_1$</th>
<th>$B_{1,1}$</th>
<th>$A_{1,1}$</th>
<th>$B_{2,1}$</th>
<th>$A_{2,1}$</th>
<th>$B_{2,1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_x$</td>
<td>4.6576</td>
<td>0.04908</td>
<td>0.04750</td>
<td>204.2586</td>
<td>120.7984</td>
<td></td>
</tr>
<tr>
<td>$n_y$</td>
<td>4.76892</td>
<td>0.04480</td>
<td>0.05430</td>
<td>221.3730</td>
<td>134.2832</td>
<td></td>
</tr>
<tr>
<td>$n_z$</td>
<td>7.97109</td>
<td>0.06079</td>
<td>0.05062</td>
<td>1234.6913</td>
<td>269.8904</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2. (Color online) Normalized thermo-optic coefficients of RTP at 20°C. This work, open circles correspond to experimental data (Table 1) and solid curves to Eq. (6). Dotted, dashed and dotted-dashed curves represent the fitting results deduced from previous works [10,4,3]: black, blue, and red colors refer to $X$, $Y$, and $Z$ polarizations, respectively.

Fig. 3. (Color online) Doubling efficiency normalized to the largest peak value recorded at high temperature, as a function of temperature and for various selected propagation angles $\varphi$ in the $(X,Y)$ plane of RTP and for a crystal length $L = 5.2$ mm. Colored open triangles, circles, stars, kozenges, and squares are experimental data obtained at $\varphi = 56.45^\circ$, 56.83°, 60.0°, 66.39° and, 66.70°, respectively. Solid curves drawn in same black, red, blue, violet, and green colors represent the theoretical predictions. The dashed curve corresponds to the optimized configuration for $L = 5.2$ mm, which occurs at $T_m = 95^\circ$C and $\varphi_m = 56.33^\circ$.
Fig. 4. (Color online) Experimentation analogous to that of Fig. 3, performed on a sample of 15 mm in length. Colored open squares, stars, and circles are experimental data obtained at $\varphi = 55.59^\circ$, $55.61^\circ$, and $55.40^\circ$, respectively. Solid curves drawn in same, red, blue, and black colors represent the theoretical predictions. For illustration, solid curve in bold open black circles displays the optimized configuration calculated for a conventional crystal of 10 mm in length.

B. Theoretical Analysis

1. Temperature Dependence of the Phase-Matching Angle

Type II SHG 1.0642 $\mu$m $\rightarrow$ 0.5321 $\mu$m is governed by the phase mismatch $\Delta k$ at the fundamental wavelength $\lambda_0$, written as in the $(X, Y)$ plane of the dielectric frame:

$$\Delta k(\lambda_0, T, \varphi) = \frac{2\pi}{\lambda_0} [2n(\lambda_0/2, T, \varphi) - (n_1(\lambda_0, T, \varphi) + n_2(\lambda_0, T))].$$

(9)

where $\lambda_0 = 1.0642 \mu$m, $n(\lambda_0/2, T, \varphi)$ and both $n(\lambda_0, T, \varphi)$ and $n_2(\lambda_0, T)$ are the temperature- and orientation-dependent refractive indices at the second harmonic and fundamental waves, respectively. For an internal propagation angle $\varphi$, the refractive index $n$ is given by

$$\frac{1}{n^2(\lambda_0, T)} = \cos^2 \varphi \frac{1}{n_2^2(\lambda_0, T)} + \sin^2 \varphi \frac{1}{n_2^2(\lambda_0, T)}.$$  

(10)

Type II SHG phase-matched interaction in the $(X, Y)$ plane corresponds to $\Delta k = 0$, which is for

$$n(\lambda_0/2, T, \varphi) = \frac{1}{2} [n_1(\lambda_0, T, \varphi) + n_2(\lambda_0, T)].$$

(11)

In birefringent RTP and starting from Eq. (1), this occurs in the $(X, Y)$ crystallographic plane at $T_0 = 20^\circ \text{C}$ for $\varphi_{\text{opt}} = \varphi_0 = 55.82^\circ$. The temperature dependence of $\varphi_{\text{opt}}$ is determined by solving Eq. (11) numerically through a suitable procedure [16] and by using the $n_1(\lambda, T)$ values derived from our NTSC's measurements as described in Section 2. The result is drawn as a solid curve in Fig. 5, which gives the predicted evolution of the phase-matching angle $\varphi_{\text{opt}}$ from $-20^\circ \text{C}$ up to $230^\circ \text{C}$, along with the dashed curve that can be deduced from [11]. A clear parabolic behavior is observed, well fitted by the equation $\varphi_{\text{opt}} = 55.50706 + 0.01762T - 9.184 \times 10^{-5}T^2$; the phase-matching angle increases with temperature from $-20^\circ \text{C}$ up to about $100^\circ \text{C}$ and then drops for higher temperatures. Thus, we observe for the first time to our knowledge that an extremum in the temperature evolution of the type II phase-matching direction occurs near $100^\circ \text{C}$ for the SHG of Nd:YAG lasers in the $(X, Y)$ plane of RTP. One can notice also that experimental determination of phase-matching maxima recorded on Figs. 3 and 4 is in very good accordance with the theoretical prediction. This result is in strong disagreement with the thermo-optic formulas proposed in [11]: the corresponding dashed curve drawn in Fig. 5 predicts a continuous decrease of the phase-matching angle when increasing the temperature, and two peaks of efficiency would never occur in the vicinity of $\varphi = 56^\circ$, in contrast to what we observed.

2. Thermal Evolution of Doubling Efficiency

It is clear from experimentation that the classical meaning of "thermal acceptance," expressed in °C cm units and usually taken as the FWHM of a single and symmetric sinc$^2$ function, does not apply in the case of RTP. To describe the thermal behavior, we will express the phase mismatch of Eq. (9) for a given change in temperature $\Delta T$ from $T_0$ and for a given internal tilt angle $\delta \varphi$ around the initial value $\varphi_0$. Experimentally, $\delta \varphi$ is deduced from the chosen external tilt angle $\Delta \varphi$ from normal incidence through the relationship $\delta \varphi = \Delta \varphi / 0.5[n(\lambda_0, \varphi_0) + n_2(\lambda_0)]$ [17].

In a low-energy regime, the efficiency dependence on phase mismatch is proportional to

$$\frac{d \text{eff}}{d \varphi} \propto \frac{\sin^2[\Delta k(\lambda_0, T, \varphi) \cdot L(T)/2]}{n(\lambda_0/2, T, \varphi) \cdot n_1(\lambda_0, T, \varphi) \cdot n_2(\lambda_0, T)} \cdot \frac{[\Delta k(\lambda_0, T, \varphi) \cdot L(T)/2]^2}{[\Delta k(\lambda_0, T, \varphi) \cdot L(T)/2]^2},$$

(12)

d$\text{eff}$ is the effective nonlinear coefficient, $L(T)$ is the crystal thickness and $\Delta k(\lambda_0, T, \varphi)$ is given by Eq. (9). For an internal tilt angle $\delta \varphi = \varphi - \varphi_0$ and a temperature variation $\delta T = T - T_0$, the phase mismatch may be expanded to the first order as

Fig. 5. Predicted temperature dependence of the phase-matching angle in the $(X, Y)$ plane of RTP for the SHG of Nd:YAG lasers. Solid curve, this work, along with open circles representing the polynomial fit (see Subsection 3.B.1); the dashed curve figures the result deduced from [11]. Black squares correspond to experimental maxima of efficiency recorded on both Figs. 3 and 4.
\[ \Delta k(\lambda_0, T, \varphi) = \Delta k_{\text{rev}} + \frac{\partial \Delta k}{\partial \varphi} \bigg|_{\lambda_0, \varphi} \delta \varphi \]

\[ + \frac{\partial}{\partial T} \left[ \Delta k_{\text{rev}} + \frac{\partial \Delta k}{\partial \varphi} \bigg|_{\lambda_0, \varphi} \delta \varphi \right] \delta T, \]  
\]

(13)

knowing that \( \Delta k_{\text{rev}} = 0 \).

From Eq. (10), we can write the following two derivatives:

\[ \frac{\partial n(\lambda_0)}{\partial \varphi} = \frac{1}{2} \left[ \frac{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi}{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi} \right]^{3/2}, \]  

(14)

\[ \frac{\partial n(\lambda_0)}{\partial T} = \left[ \frac{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi}{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi} \right]^{3/2} \left[ \frac{3n_y(\lambda_0) \sin \varphi \cos \varphi + n_y(\lambda_0) \sin \varphi \cos \varphi}{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi} \right]. \]  

(15)

Equation (13) may be written as

\[ \Delta k(\lambda_0, T, \varphi) = \Delta k_{\text{rev}} + \frac{\partial \Delta k}{\partial \varphi} \bigg|_{\lambda_0, \varphi} \delta \varphi + \frac{\partial \Delta k}{\partial T} \bigg|_{\lambda_0, \varphi} \delta T \left[ \frac{3n_y(\lambda_0) \sin \varphi \cos \varphi + n_y(\lambda_0) \sin \varphi \cos \varphi}{n_x^2(\lambda_0) \cos^2 \varphi + n_y^2(\lambda_0) \sin^2 \varphi} \right]. \]  

(16)

The phase mismatch given by Eq. (9) can then be calculated from Eq. (16) by using Eqs. (14) and (15) along with our definition of NTOCs, accounting for the respective following expressions which are to be taken at \( T = T_0 \) and \( \varphi = \varphi_0 \).

In Eqs. (17)–(19) we have used for convenience the subscripts \( e \) and \( 2e \), which are the pulsations corresponding to wavelengths \( \lambda_0 \) and \( \lambda_0/2 \), respectively.

In Eq. (12) we also account for the change in sample length with temperature; it can be shown that it is given by the following expression:

\[ L(T) = L(T_0) \times \exp \int_{T_0}^{T} (a_x \cos^2 \varphi_0 + a_y \sin^2 \varphi_0) dT. \]  

(20)

Moreover, for a given propagation angle, we noticed that the main \( \sin^2 \Delta k L/2 \) peak power occurring at high temperatures is slightly more intense than the one observed at lower ones. For instance, in Fig. 3, maxima of efficiency are located at \( T_M = 169.5^\circ \text{C} \) and \( 167.5^\circ \text{C} \) for \( \varphi = 56.66^\circ \) and \( 55.86^\circ \), respectively, and at \( T_M = 163^\circ \text{C} \) for \( \varphi = 55^\circ \). For \( \varphi = 55.00^\circ \) in Fig. 4. Corresponding secondary maxima occurring at lower level are situated at \( 41^\circ \text{C} \), \( 23^\circ \text{C} \), and \( 27^\circ \text{C} \), respectively. To account for this behavior, we considered that the conversion efficiency is weighted by the coefficient \[ d_{\text{eff}}(T) = \frac{d_{\text{eff}}(T)}{d(\lambda_0, T, \varphi)} \] (see Eq. (13)). Since the variation of the product \( n(\lambda_0)/2, T, \varphi \cdot n(\lambda_0, T, \varphi) \cdot n(\lambda_0, T) \) is not significant over the explored temperature interval, we assumed that the nonlinear \( d_{\text{eff}} \) optical coefficient was also temperature dependent and found that, for all curves, best fits of efficiency normalized to the highest peak value were obtained by using the quadratic development:

\[ d_{\text{eff}}(T) = d_{\text{eff}}(T) \left( 1 + 1.3116 \times 10^{-4}(T - T_M) + 5 \times 10^{-7}(T^2 - T_M^2) \right). \]  

(21)

\( T_M \) is the high temperature at which phase matching occurs, with a corresponding value \( d_{\text{eff}} \) of the effective NLO coefficient. The \( d_{\text{eff}} \) values plotted versus temperature in Fig. 5 were calculated from Eq. (22) and experimental data of \( T_M \), taking \( d_{\text{eff}} = 2.6 \pm \text{pm}^2 \nu \) at \( 20^\circ \text{C} \) [5]. A least-square fitting procedure gave the following expression of the temperature dependence of the effective NLO coefficient, drawn as a dotted curve in Fig. 6:

\[ d_{\text{eff}} = 2.597 \times 1.406 \times 10^{-4} T + 7.19 \times 10^{-7} T^2. \]  

(22)

We noticed that such an increase in the value of the second-order NLO coefficient with increasing temperature has been already analyzed in the case of LiNbO₃ [18].

4. DISCUSSION

A. Phase-Matching Angle

In Subsection 3.B.1 we have seen that the measured temperature dependence of the type II phase-matching angle \( \varphi \) in the critical \( (X, Y) \) plane of RTP is very well predicted starting from Eq. (11), along with the methodology employed to determine the dispersion of NTOCs. The most striking feature is
that an extremum in the evolution of $\phi$ occurs near 100°C, which cannot be explained by the formulation proposed previously [11].

Measurements of refractive indices have been early reported in [13]. A one pole Sellmeier equation along with a single IR correction proportional to $x^2$ was proposed, from which we have deduced the corresponding dispersion of NTOCs starting from the experiment summarized in Table 2.

Of course, in that case measurements at four wavelengths only are sufficient for their entire determination, and there was no need to assume any temperature dependence of a nonexistent IR pole. They also performed measurements of thermal acceptance for type II SHG of a YAG laser in the $(X, Y)$ plane in a similar way as we did, namely by tilting a crystal of 12.4 mm in length from an (unspecified) angle $\Delta \phi$ from the configuration chosen for maximum doubling efficiency at room temperature. We have reproduced at the same scale their experimental results in open circles in Fig. 7, along with our theoretical analyses.

In the latter case, the phase-matching angle at $T = 20$°C in the $(X, Y)$ plane is 56.8° and not 58° as claimed by authors. We thus estimate that the (internal) tilt angle leading to the data in Fig. 7 was $\Delta \phi = 0.45^\circ$, corresponding to an external value $\Delta \phi = n_{ex} - n_{in}$, where $n_{ex} = 0.5(i \lambda_0 \phi_0) + n_{in}$. For the determination of the temperature dependence of the phase-matching angle, both Sellmeier equations gave quite similar results, as evidenced in Fig. 8. We notice also that such agreement confirms that the temperature dependence of the IR pole included in the Sellmeier formulas of [11] is insignificant and justifies the assumption $dE/d\alpha = 0$ mentioned in Subsection 2.B.2. The dispersion formula proposed in [12] showed an evolution that was out of scale ($\phi_{0\alpha} = 59.7$° at 20°C), far from experimental observations.

B. Temperature Bandwidth of the SHG

In contrast to the earlier statement [13], experimental results and modeling displayed in Figs. 3 and 4 clearly show that no temperature acceptance can be assigned to critical type II SHG in the $(X, Y)$ plane of RTP without specifying the internal propagation angle $\phi$ referred to the $X$ axis as well as the operating temperature. The conversion efficiency depends strongly on both coupled parameters. From a practical point of view, an interesting feature consists in establishing the conditions for which phase-matching may be obtained with a temperature bandwidth as wide as possible. From Eq. (12) and using the dispersion formula of [11], we determined that this is achieved for $\phi_M = 56.305^\circ$ and $T_M = 95^\circ$C, with a FWHM temperature acceptance $\Delta T = 117^\circ$C for a crystal of length $L_M = 10$ mm. The curve corresponding to this peculiar case is drawn in open circles in Fig. 4. The modeling shows that, in this configuration, a single and quasi-symmetric peak of efficiency occurs, with a plateau extending from about 75°C up to 120°C. Inside this interval, the crystal may be said to be temperature insensitive for type II SHG. As an illustration, we can see that the experimental data corresponding to $\phi = 56.30^\circ$ in Fig. 3 are quite close to this case, the modeling of which being also displayed as a dotted curve for $L = 5.2$ mm.

C. Case of ppRTP

Temperature tuning of ppRTP optical parametric oscillators with various QPM periods as well as phase-matched grating
Fig. 9. Temperature dependence of the phase-matched grating periods for the SHG at 0.5321 μm with Nd:YAG laser pump wavelength. Open circles, experimental results [4]; solid, dashed, and dotted curves are the fits obtained starting from available dispersion equations reported in [11–13], respectively. Corresponding NTOCs were calculated in each case.

periods as a function of crystal temperature in SHG at 0.5321 μm with Nd:YAG laser pump wavelength has been reported previously [3,4]. In the last case the fundamental beam propagates along the X axis with a polarization state oriented along the Z one. As an extension to the study performed on bulk crystals, we attempt to fit the experimental results given for the SHG in ppRTP by using thermal expansion and NTOC values given in Section 2; the result is displayed as a solid curve in Fig. 9.

A quasi-constant discrepancy between experimental results and the solid curve is observed, which does not exceed 0.05 μm over the 30°C–120°C temperature interval. At such scale our modeling appears quite satisfactory since it could lie inside the error bar that may be thought associated to the experimental determination of the grating periods. We notice also better adjustment to experimental values by applying our formalism starting from the dispersion equation of [11].

Better fits could have been obtained by using different formulas for the dispersion of thermo-optic coefficients [4,11]. However, in contrast to our analysis, the fitting procedures were based on a development of dnt/dT in a power series of 1/λ, which is not in accordance mathematically with the Sellmeier equations from which they should have been derived.

5. CONCLUSION

Interferometric determination of thermal expansion and of NTOCs were performed from −30°C up to +130°C on parallelepipedic and millimetric-sized RTP crystals cut with respect to the X, Y, and Z principal axes. Four laser wavelengths at 0.4578, 0.6328, 1.0642, and 3.39 μm were used to obtain an analytical expression of the temperature dependence of the dispersion of NTOCs, which further allowed the establishment of Sellmeier equations of refractive indices as function of both wavelength and temperature, starting from appropriate dispersion formulas given for Tg = 20°C.

Using the chosen angle, two maxima of efficiency may appear at low and high temperature, respectively. Subsequent theoretical analysis performed from our determination of nT(T) predicts a quadratic behavior of phase-matched interaction with temperature, in very good accordance with experimental data. It is shown also that these two maxima collapse into a single one for θ = 56.33° at T = 100°C; in these conditions, an important consequence is that the largest FWHM temperature bandwidth can be obtained, with a value of 117°C for a sample of 10 mm in length. Any other condition for the couple of T and θ values leads to much lower and non-symmetric FWHM, due to the quadratic temperature dependence of NTOCs; this fully explains the so-called "enigmatic" discrepancy between measurements of the phase-matching temperature acceptance bandwidth of the SHG of Nd:YAG lasers in the (X, Y) plane of RTP [19]. Moreover, we can outline that heating a doubler up to 100°C for the best condition of SHG appears also very appropriate in the meantime to overcome the drawback of an eventual occurrence of photorefractive effects.

Despite having determined the NTOCs only from −30°C up to 130°C, the good agreement between measurements and modeling of the thermal evolution of conversion efficiency carried out up to 220°C indicates that our dispersion formulas of thermo-optic coefficients are also valid above 130°C. We notice also that a slight quadratic temperature dependence of the dnt NLO coefficient had to be assumed to achieve best adjustments of theoretical analysis to experimental data.

ACKNOWLEDGMENTS

Thanks are due to P. Strimer and Dr. S. Fossieler-Passaguil for their help in thermal expansion and thermo-optic coefficients measurements.

REFERENCES


