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Temperature dependence of $LiTaO_3$ refractive index: Corrections of Sellmeier equation

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Abstract. We report a new and more precise Sellmeier equation obtained by using the analysis of quasi-phase-matching curves of the optical parametric generation (OPG) in 1D periodically poled $LiTaO_3$ (1D-PPLT) of different grating periods.

Keywords: Sellmeier equation, Lithium tantalate, Nonlinear photonic crystals, Nonlinear optics.

1 Introduction

Lithium tantalate ($LiTaO_3$) possesses unique electro- optical, pyroelectric and piezoelectric properties combined with good mechanical and chemical stability, a high optical damage threshold [1], high resistance to photo-refractive effects, high non-linear coefficients [2], and a broad transparency range spanning from 280 to 5500 nm allowing for frequency conversion ranging from UV to infrared [3, 4]. These characteristics render it highly suitable for numerous applications, particularly in the field of nonlinear optics, makes it a very interesting material for the realization of non-linear periodically polarized components [5] including electro-optical modulators, pyroelectric detectors, optical waveguides and SAW substrates, piezoelectric transducers, etc. [6, 7].

$LiTaO_3$ is a nonlinear positive uniaxial crystal with low birefringence and belongs to the 3 m (C_{3V}) trigonal crystallographic group [8]. The elements of its second-order tensor $\chi^{(2)}$ allow for three more types of nonlinear interactions: o-oo (d_{22}, d_{21}, d_{16}), e-oo (d_{31}, d_{32}), and o-eo (d_{24}, d_{15}) [8]. Because of the highest nonlinear susceptibility tensor element d_{33} ~ 16 pm/V, the most commonly used nonlinear interaction is e-ee, where an extraordinary wave generates two other extraordinary waves. In addition, for these interactions, only the extraordinary index is needed [9].

Accurate knowledge of the dispersion of the extraordinary refractive index is crucial for designing frequency conversion devices as well as interpreting experimental results of nonlinear interactions. Typically, an accuracy exceeding 10^{-4} of the refractive index is needed to correctly predict phase matching terms of frequency conversion processes [8].

Various techniques are employed to measure refractive index variations [10]: spectrophotometry, ellipsometry, m-lines, and minimum deviation can yield measurements with accuracies ranging from ± 0.05 to $\pm 10^{-3}$.

Besides, directly deducing the Sellmeier equation from experimental phase matching curves [11] offer an accuracy greater than 10^{-4} and makes this technique highly effective.

Different processes are reported such as the sphere method developed by Boulanger *et al.* [11].

Another method is to deduce the Sellmeier equation from the curves of the quasi-phase matching obtained by characterizing samples of several periods by varying the pump wavelength [12] or varying the temperature [13–15]. Indeed, a simultaneous interpolation of all the quasi-phase matching curves measured, allows to fit all the coefficients of the Sellmeier equation and thus, to find out a very precise index dispersion of the crystal studied [5, 11]. It should be noted that $LiTaO_3$ exists under different types: stoichiometric (SLT), congruent (CLT), and doped stoichiometric. Several investigations have reported the Sellmeier equation for the various lithium tantalate types. However, as far as our knowledge extends, only three versions of the Sellmeier equation have been reported in the literature specifically for the CLT type [13–15]. In 1996, Abedin *et al.* [13] defined the Sellmeier equation of $LiTaO_3$-CLT using the experimental measurement. Then, Mayen *et al.* [14] and Bruner *et al.* [15] derived this equation from the phase matching curves.

While the existing studies [13–15] concern the entire transparency range of $LiTaO_3$ and cover a sufficient temperature interval, we recently showed a significant discrepancy between these different Sellmeier equations and theoretical and experimental measurements, as well. Moreover, when encountering several nonlinear phenomena

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in the same sample, especially at high power pumping, the interpretation using these established Sellmeier equations proved challenging [16].

In this work, we propose a novel and more accurate Sellmeier equation derived from quasi-phase-matching curves obtained from the investigation of optical parametric generation (OPG) in 1D periodically poled $LiTaO_3$ (1D-PPLT) crystals with varying periods.

Sellmeier equation which contains the dependence of the refractive index on temperature is essential to consider thermos-optics effect when developing functional optical systems, particularly in applications where stability and control of optical properties are crucial, such as frequency generation devices, sensors, and stabilized lasers.

2 Experimental procedure and method

In order to study the optical parametric generation (OPG) process in PPLT crystals, we used the experimental setup, which its simplified scheme is depicted in Figure 1.

We used the one dimension periodically poled $LiTaO_3$ (1D-PPLT) nonlinear photonic crystals fabricated by the electric poling technique [17]. The investigated grating periods Λ include 8.08 μm, 8.29 μm, 8.43 μm, 8.52 μm and 12 μm. The filling ratio is 50%, chosen to obtain the most efficient frequency conversion [18]. The samples were made on z-cut congruent-grown $LiTaO_3$ with dimensions of 1.5 (W) × 2 (L) × 0.5 (thickness) cm^3.

The samples are pumped by a Q-switch doubled Nd: YAG pump laser frequency doubled at 532 nm with a pulse duration of 0.5 ns. The pulse repetition rate can be adjusted from 10 Hz to 1 kHz. A half-wave plate is used to control the polarization of the pump beam, which is aligned with the PPLT z-axis in order to take advantage of the crystal largest nonlinear coefficient d_{33}. The samples are placed on a temperature controller, allowing the crystal temperature to be adjusted within a range of 40 °C to 200 °C with an accuracy of ±0.1 °C. The optical parametric generated (OPG) beam is coupled to a 50 μm optical fiber using an optical lens followed by microscope objective. The optical fiber is connected to an optical Spectrum Analyzer (OSA, ANDO AQ6315A), which can take measurement with a spectral coverage from 350 nm to 1750 nm with a resolution between 0.05 nm and 10 nm. A 532 nm stop-band filter was used to absorb the residual output pump beam.

For each sample, we measured the corresponding OPG spectra with a resolution of 0.5 nm. The measurements were performed over a temperature range from 40 °C to 200 °C, with increments of 2 °C, and a pump energy of 15 μJ.

Subsequently, we developed a specific Matlab code capable of simultaneously fitting all the measured OPG spectral data corresponding to a temperature ranging from 40 °C to 200 °C for a give QPM period in the PPLT samples. This code is employed to derive the Sellmeier equation which, together with the grating period of the PPLT sample, fulfils the conservation of momentum for the pump, the signal, and the idler wavelength in the basic quasi phase matching (QPM)- OPG conditions as follows.

For fulfilling the law of energy conservation, the wavelengths of the pump, the signal, and the idler of λ_p, λ_s, λ_i obeys:

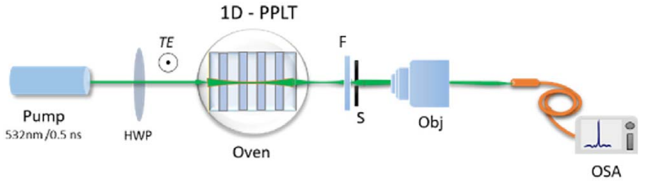

Figure 1. Scheme of the experimental setup utilized for the optical characterization of nonlinear photonic crystals.

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i} \tag{1}$$

The conservation of momentum expressed by the QPM-OPG condition for a given PPLT periodicity of Λ is:

$$\frac{n_{ep}(\lambda_p, T)}{\lambda_p} - \frac{n_{es}(\lambda_s, T)}{\lambda_s} - \frac{n_{ei}(\lambda_i, T)}{\lambda_i} - \frac{m}{\Lambda(T)} = 0 \tag{2}$$

Where $\lambda_{p,s,i}$ denote the wavelengths of the pump, signal and idler. $n_{e,p,s,i}$ represent the extraordinary refractive indices corresponding to the pump, signal and idler, respectively. m is an integer that defines the order of the quasi-phase matching, and Λ signifies the period of the lattice.

Additionally, we consider the thermal expansion effect of the lithium tantalate lattice, as described by Y.S. Kim *et al.* [19].

$$\begin{aligned}\Lambda(T) = \Lambda(20^\circ\mathrm{C})[1 + 1.6\times10^{-5}(T - 20^\circ\mathrm{C}) \\ + 7\times10^{-9}\left(T - 20^\circ\mathrm{C}\right)^2\Big]\end{aligned} \tag{3}$$

The used Sellmeier equation model is based on the formulation reported by [7, 8]:

$$n_e^2(\lambda, T) = A + \frac{B + b(T)}{\lambda^2 - \left(C + c(T)^2\right)} + \frac{E}{\lambda^2 - F^2} + D\lambda^2 \tag{4}$$

A, B, C, D, E and F are constant parameters, b and c are coefficients that vary with temperature. The wavelength λ is expressed in micrometers.

For determining the latter formulation, we consider the OPG-(signal, idler) wavelength data obtained from the five sets of PPLT samples of QPM periodicity Λ in 8.08 μm, 8.29 μm, 8.43 μm, 8.52 μm and 12 μm, respectively, over a temperature range from 40 to 200 °C. We, then, apply equation (4) to retrieve the corresponding coefficients from A to F for the temperature dependent Sellmeier Eq. with our Matlab coding subject to the least square method. In writing the Matlab codes to solve the Sellmeier equation, we have taken into account the QPM-OPG processes and considered the lattice thermal expansion effect [19].

3 Results and discussions

As an example, the OPG spectra, recorded at $T = 110$ °C and a pump energy of 15 μJ for 1D-PPLT with a period $\Lambda = 8.29$ μm shown in Figure 2(a), reveal two peak wavelengths at (851, 1418) nm which corresponds to the signal and idler, respectively. The mapping of signal and idler

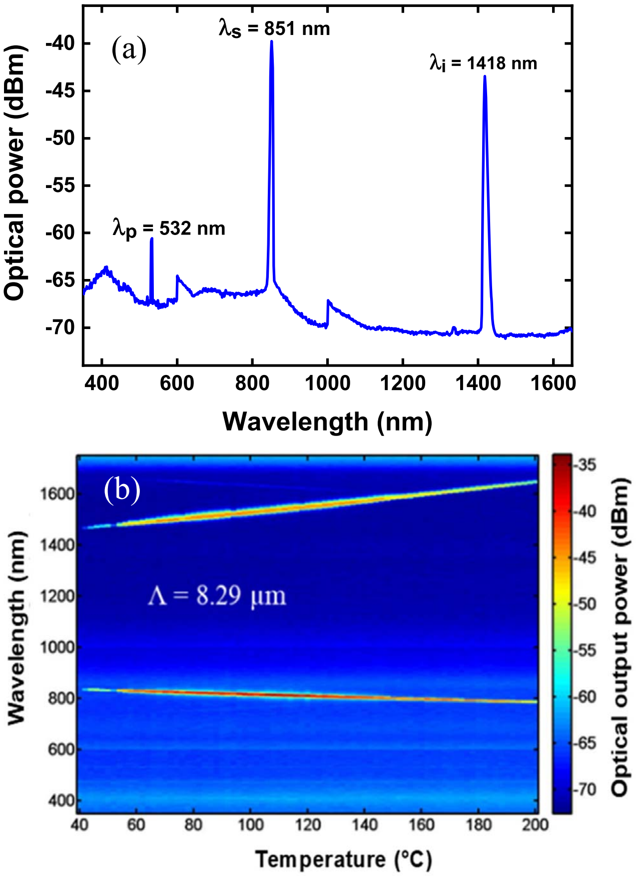

Figure 2. (a) Spectrum of the signal (851 nm) and idler (1418 nm) measured at 110 °C. (b) OPG map from 40 °C to 200 °C generated from 1D-PPLT of Λ = 8.29 µm at 15 µJ.

wavelengths generated by the OPG process as a function of temperature ranging from 40 °C to 200 °C are represented in Figure 2(b).

Note that for the 12 µm period PPLT sample, the spectral detection limits of the OSA prevent to record the idler waves in the mid-infrared (3 µm) spectral regime. Thus, the corresponding idler wavelengths, between 3.355 µm and 3.67 µm, are calculated from the measured signal waves.

For instance, Figure 3. displays the variation of the signal wavelength as a function of temperature for 1D-PPLT samples of Λ = 8.52 µm and Λ = 12 µm, respectively.

Our experimental data are compared to those calculated by using the three Sellmeier models already reported [13–15]. It is worth noting that similar results have been obtained in the case of the idler.

It is evident that the experimental signal wavelengths differ from those calculated by the existing formulae of Sellmeier equations cited in references [13–15]. This discrepancy is more pronounced when using the equation proposed by Abedin *et al.* [13], where the wavelength of OPG-signal/idler deviates significantly with the temperature increase. This variation can be attributed to the optical reflection measurement technique used to retrieve the raw refractive index data cited in [13] to define the Sellmeier equation. The divergence is less pronounced with the predictions made by the other two variants of Sellmeier Eq. in [14] and [15].

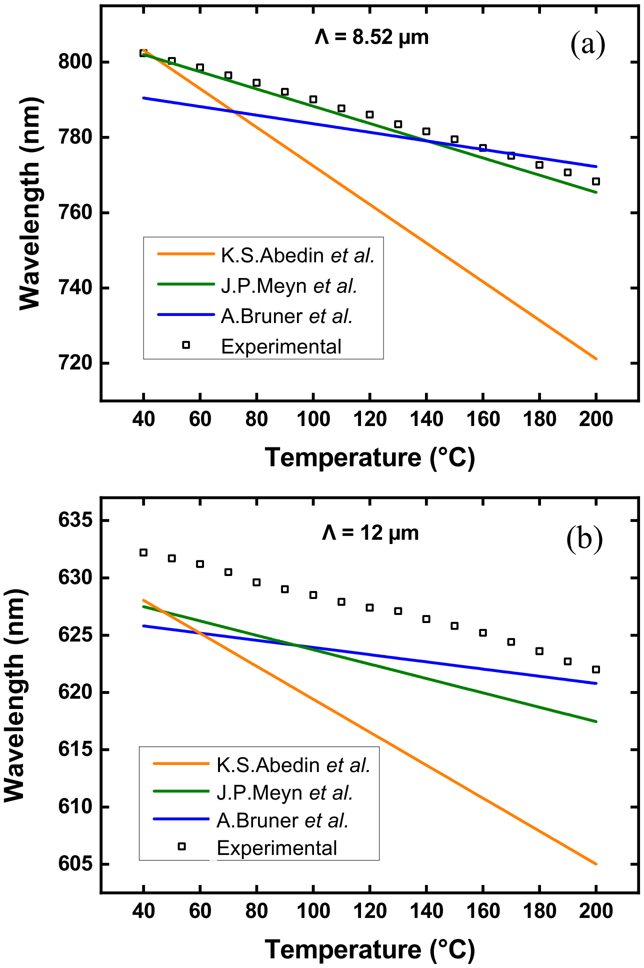

Figure 3. Signal wavelengths vs. temperature for 1D-PPLT of (a) Λ = 8.52 µm and (b) Λ = 12 µm.

Although the discrepancies between theoretical and experimental results may be attributed to various sources of errors, such as pump and temperature fluctuations, possible irregularities in the periodic lattice, equipment precision, and measurement errors, the observed divergences are significant and increase with the rise in temperature and period. For instance, using the equation provided by Meyn *et al.*, a variation of the period ranging from 0.004 µm to 0.03 µm is observed when the temperature varies from 40 °C to 200 °C for the sample of a period of 8.52 µm. A substantial deviation (0.2 µm at 200 °C) is noted for the sample with a period of 12 µm.

Again, these results emphasize the importance of precisely determining the Sellmeier coefficients.

Finally, we analyzed the results obtained from the study of the five sets of 1D-PPLT samples of QPM periodicity Λ in 8.08 µm, 8.29 µm, 8.43 µm, 8.52 µm and 12 µm, as reported on Figure 4. for the signal (note that similar work was performed in the case of the idler, as well).

The experimental data were fitted using the same Sellmeier equation, allowing us to determine the best results

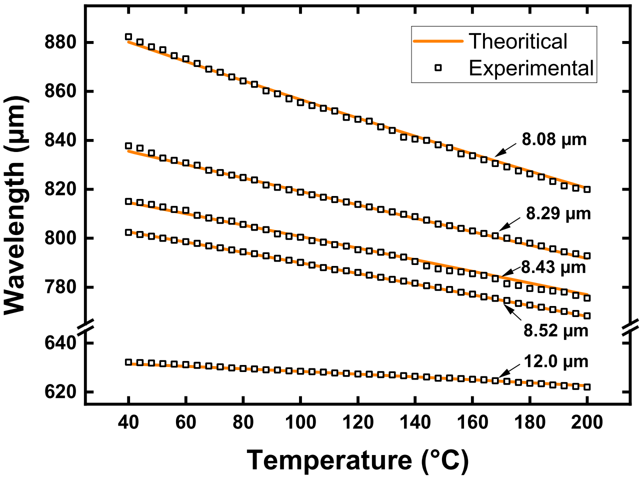

Figure 4. Theoretical (red line) and experimental Signal wavelengths (squares) as a function of the temperature for 1D-PPLT Λ = 8.08 µm, 8.29 µm, 8.43 µm, 8.52 µm and 12 µm.

Table 1. Sellmeier equation parameters verifying the QPM wavelengths over the range 0.6–3.6 µm and 40–200 °C.

A	4.5281
B	0.00724542
C	0.2439
D	−0.02172
E	0.07858
F	0.1835
b(t)	$2.5488 \times 10^{-8}T^2$
c(t)	$1.6225 \times 10^{-8}T^2$

obtained giving the new coefficients of the Sellmeier equation are as reported in Table 1.

Note that the fitting process involves both the signal and the idler. The resulting equation is valid for wavelengths between 0.6 µm and 3.6 µm and for a temperature range between 40 and 200 °C.

It is important to indicate that the above parameters have different influences. For example, unlike parameter A, a small variation in parameter B results in a significant change in n.

To confirm these results, we first conducted a comparative study between the experimental results and theoretical simulations using the new equation.

To illustrate the validity of the proposed equation, we compared the effective periods of the studied samples with those obtained from the different Sellmeier equations. Because the period is a physical property given by the fabrication procedure, we used the different equations to calculate the periods of the 5 1D-PPLT gratings studied, from the OPG measurements. The results obtained show significant divergences as indicated in Figure 5. In fact, this figure reports the difference between the calculated values and those given by the fabrication technique (named period error). However, this difference is negligible when using the equation that we propose in this work. This further confirms the validity of this new equation.

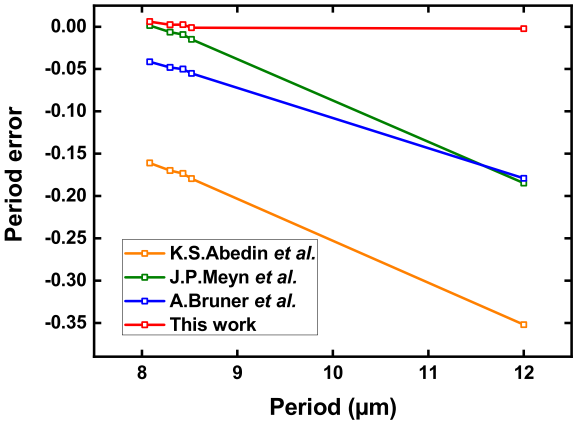

Figure 5. The period error at T = 110 °C.

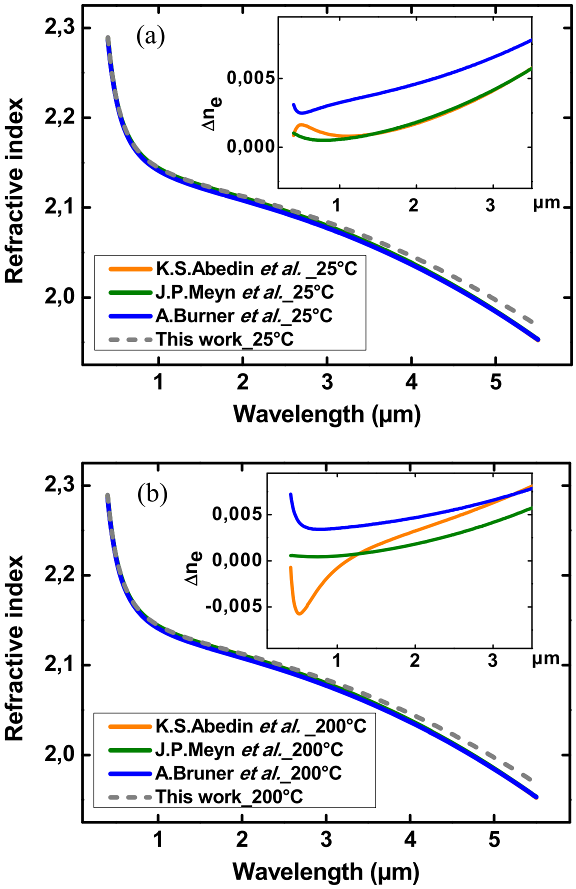

Figure 6. Comparison of the extraordinary refractive index dispersion n_e as a function of wavelength obtained using the previous Sellmeier equations and the new proposed equation. (a) T = 25 °C and (b) T = 200 °C. The zooms represent the difference of refractive index values obtained with the new equation and the previous ones.

To go further in the analysis of the validity of our new corrected sellmeier equation related to already published ones, we present in Figure 6 the plot of dispersion curve for n_e with wavelength at room temperature and 200 °C, respectively.

Considerable deviations are observed between the refractive indices plotted using different Sellmeier equations compared to the one proposed in this work. The most significant differences are obtained with the refractive index calculated from the equation given by Bruner *et al.* [15], and this holds across the entire wavelength range for both $T = 25$ °C and 200 °C. At low temperatures, the difference increases with the increase of wavelength, especially for wavelengths close to the infrared region. The calculated error of the extraordinary refractive index ranges between -6.10^{-3} and 7.10^{-3}. These discrepancies are substantial enough to introduce errors in the design of optical devices.

4 Conclusion

Our exploration of optical parametric generation with respect to temperature variations in PPLT-1D crystals with various periods has enabled us to identify and present a more precise Sellmeier equation. The chosen criterion was associated with the temperature-dependent evolution of the grating periods in the samples. The refined equation provides a more accurate extraordinary refractive index for congruent lithium tantalate.

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Conflicts of interest

We declare that we have no conflicts of interest to report.

Data availability statement

All the data used in this paper are available upon request.

Author contribution statement

Safia Mohand Ousaid performed the experimental work and the analysis of the obtained results as this is a part of her PhD project.

Kai H. Chang worked in close connection with Safia Mohand Ousaid, specially for the analysis of the results obtained.

Professors Lung Han Peng and Azzedine Boudrioua as the PhD supersivors, provided their expertise in conducting the work reported in this paper as well as in the preparation of the paper and the corrections.

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